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Zhang, Yifeng

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DTU Environment
Department of Environmental Engineering

PhD Thesis
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Address: DTU Environment
Department of Environmental Engineering
Technical University of Denmark
Miljoevej, building 113
DK-2800 Kgs. Lyngby
Denmark

Phone reception: +45 4525 1600
Phone library: +45 4525 1610
Fax: +45 4593 2850

Homepage: http://www.env.dtu.dk
E-mail: reception@env.dtu.dk

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Preface

The work reported in this PhD thesis, entitled "Energy recovery from waste streams with microbial fuel cell (MFC)-based technologies", was conducted at the Department of Environmental Engineering at the Technical University of Denmark from October 2009 to September 2012. Professor Irini Angelidaki was the supervisor.

The thesis is organized in two parts. The first part is an introductive review and summary; the second part is constituted by the papers published or going to be submitted to scientific journals.

In the text, the papers shown below are referred to by names of the authors and their appendix number written with roman numbers.

I. Zhang Y., Noori J.S., Angelidaki I. 2011. Simultaneous organic carbon, nutrients removal and energy production in a photomicrobial fuel cell (PFC). *Energy and Environmental Science* 4(10): 4340-4346.

II. Zhang Y., Angelidaki I. 2012. Innovative self-powered submersible microbial electrolysis cell (SMEC) for biohydrogen production from anaerobic reactors. *Water Research* 46(8): 2727-2736.

III. Zhang Y., Angelidaki I. 2011. Submersible microbial fuel cell sensor for monitoring microbial activity and BOD in groundwater: Focusing on impact of anodic biofilm on sensor applicability. *Biotechnology and Bioengineering* 108(10):2339-2347.

IV. Zhang Y., Angelidaki I. 2012. A simple and rapid method for monitoring dissolved oxygen in water with a submersible microbial fuel cell (SBMFC). *Biosensors and Bioelectronics* 38(1):189-194.

V. Zhang Y., Angelidaki I. 2012. Self-stacked submersible microbial fuel cell (SSMFC) for improved remote power generation from lake sediments. *Biosensors and Bioelectronics* 35(1): 265-270

VI. Zhang Y., Angelidaki I. 2012. Bioelectrode-based approach for enhancing nitrate and nitrite removal and electricity generation from eutrophic lakes. Moderate revision in *Water Research.*

VII. Zhang Y., Angelidaki I. 2012. A new method for *in situ* nitrate removal from groundwater using submersible microbial desalination-denitrification cell (SMDDC). Submitted manuscript.

In addition, the following work was conducted in the period of my PhD study, while was not included in the PhD thesis.

Zhang Y., Olias L.G., Kongjan P., Angelidaki I. 2011. Submersible microbial fuel cell for electricity production from sewage sludge. *Water Science and Technology* 64(1):50-55.

Alatraktchi F.A., Zhang Y., Noori J.S., Angelidaki I. 2012. Expanding surface area of electrodes by nanotechnology to enhance electricity generation in microbial fuel cells. *Bioresource Technology*. In process, doi: 10.1016/j.biortech.2012.07.048

Alatraktchi F.A., Zhang Y., Angelidaki I. 2012. Enhanced performance of microbial fuel cells by decorating carbon paper electrodes with gold nanoparticles. To be submitted

Ucar D., Zhang Y., Angelidaki I., Cokgör E.U. 2012. An overview of terminal electron acceptors in microbial fuel cells. To be submitted

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Yifeng Zhang

The papers **I-VII** are included in the printed version of the thesis but not in the www-version.
Copies of the papers can be obtained from the
Library at DTU Environment.
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Technical University of Denmark
Miljoevej, Building 113
DK-2800 Kgs. Lyngby, Denmark
(library@env.dtu.dk)

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Just let the thesis in memory of my grandfather (Lebin Zhang)…

Thank you!

Yifeng Zhang
September 2012

There are no secrets to success.

It is the result of preparation, hard work, and learning from failure.

-Colin L. Powell

Summary

Microbial fuel cell (MFC)-based technologies are promising technologies for direct energy production from various wastewaters and waste streams. Beside electrical power production, more emphasis is recently devoted to alternative applications such as hydrogen production, bioremediation, seawater desalination, and biosensors. Although the technologies are promising, a numerous of hurdles need to be overcome before that field applications are economically feasible. The main purpose of this work was to improve the performance, reduce the construction cost, and expand the application scopes of MFC-based bio-electrochemical systems.

To reduce the energy cost in nitrogen removal and during the same process achieve phosphorus elimination, a sediment-type photomicrobial fuel cell was developed based on the cooperation between microalgae (*Chlorella vulgaris*) and electrochemically active bacteria. The main removal mechanism of nitrogen and phosphorus was algae biomass uptake, while nitrification and denitrification process contributed to part of nitrogen removal. The key factors such as algae concentration, COD/N ratios and photoperiod were systemically studied.

A self-powered submersible microbial electrolysis cell was developed for *in situ* biohydrogen production from anaerobic reactors. The hydrogen production increased along with acetate and buffer concentration. The hydrogen production rate of 32.2 mL/L/d and yield of 1.43 mol-H_2/mol-acetate were achieved. Alternate exchanging the function between the two cell units was found to be an effective approach to inhibit methanogens.

A sensor, based on a submersible microbial fuel cell, was developed for *in situ* monitoring of microbial activity and biochemical oxygen demand in groundwater. Presence or absence of a biofilm on the anode was a decisive factor for the applicability of the sensor. Temperature, pH, conductivity and inorganic solid content were significantly affecting the sensitivity of the sensor. The sensor showed good performance both with artificial and real groundwater.

A submersible microbial fuel cell sensor was developed for *in situ* and real time monitoring of dissolved oxygen (DO) in environmental waters. The current density produced by the sensor increased linearly with DO level up to 8.8±0.3 mg/L. The sensor ability was further explored under different environmental

conditions. The sensor can measure DO in different environmental waters with less deviations.

To improve the voltage output of MFC from lake sediment, an innovative self-stacked submersible MFC was developed. The system successfully produced a maximum power density of 294 mW/m^2 and had an open circuit voltage (OCV) of 1.12 V. In addition, voltage reversal was studied in detail in terms of its cause, determining parameters and elimination method. The internal resistance and OCV were the most important parameters for predicting voltage reversal. Use of a capacitor was found to be an effective way to prevent voltage reversal and at the same time store power.

A sediment-type MFC based on two pieces of bioelectrodes was employed as a novel *in situ* applicable approach for nitrate/nitrite removal, as well as electricity production from eutrophic lakes. The nitrogen removal and power generation were limited by the DO level in the water and acetate level injected to the sediment. The proposed approach may broad the application of sediment MFC technology.

A novel submersible microbial desalination cell was developed as an *in situ* and non-invasive approach for nitrate removal from groundwater. The system performance in terms of power generation and nitrate removal efficiency were investigated. The effects of hydraulic retention time, external resistance, other ionic species in the groundwater and external nitrification on the system performance were also elucidated. Over 90% of nitrate was removed from groundwater without energy input, water pressure, draw solution, additional electron donor or risk of bacteria discharge. Such a new system may offer a promising avenue for drinking water treatment and energy recovery.

Dansk Resumé

Mikrobielle brændselcelle (MFC)-baserede teknologier er lovende metoder til direkte energiproduktion fra spildevand og forskellige affaldsstrømme. Udover produktion af elektricitet er der for nyligt lagt stor vægt på at øge formålsværdien af MFC i form af brint produktion, bioremediering, afsaltning af havvand og biosensorer. Selvom teknologierne er lovende, skal talrige forhindriger overvindes før der kan være tale om økonomisk mulige feltanvendelser. Hovedformålet med dette arbejde er at forbedre ydeevnen, reducere konstruktionsomkostningerne, og udvide anvendelsen anvendelsesområder for MFC-baserede bio-elektrokemiske systemer.

For at reducere energi omkostningerne i kvælstoffjernelse og opnå fosfor eliminering under den samme proces, blev en sediment-type fotomikrobiel brændselscelle (PFC) udviklet på baggrund af samspillet mellem mikroalger (*Chlorella vulgaris*) og elektrokemisk aktive bakterier. Den største elimineringsmekanisme af kvælstof og fosfor var algebiomasse optagelse, mens nitrifikation og denitrifikations processen medvirkede til en del af kvælstoffjernelsen. De vigtigste faktorer, såsom alge koncentration, COD/N-forhold og lysperioder blev systematisk undersøgt.

En energiselvforsynende nedsænkelig mikrobiel elektrolysecelle blev udviklet til *in situ* biohydrogen produktion fra anaerobe reaktorer. Hydrogen produktionen steg sammen med acetat og buffer koncentrationen. Hydrogen produktion raten på 32,2 mL/L/d og udbytte på 1,43 mol-H_2/mol-acetate blev opnået. Alternativt viste det sig at udveksling af funktionen mellem de to celleenheder af SMEC at være en effektiv strategi til inhibering af methanogener.

En sensor, baseret på en nedsænkelig mikrobiel brændselscelle, blev udviklet til in situ mikrobiel aktivitet og biokemisk oxygenbehov i grundvandet. Tilstedeværelse eller fravær af en biofilm på anoden var en afgørende faktor for anvendelsen af sensoren. Temperatur, pH, ledningsevne og uorganisk faststofindhold påvirkede følsomheden af sensoren signifikant. Sensoren viste gode resultater både med kunstig og ægte grundvand.

En nedsænkelig mikrobiel brændselscelle blev udviklet som en biosensor for *in situ* og *real-time* overvågning af opløst ilt (DO) i miljø-farvande. Strømtætheden produceret ved sensoren øgedes lineært med DO-niveauet på op til 8,8 ± 0,3

mg/L. Sensorens evne blev yderligere undersøgt under forskellige miljøforhold. Sensoren kan måle DO i forskellige miljø-farvande med mindre afvigelser.

For at forbedre spændings outputtet af MFC fra sediment, en innovativ selv-stablet nedsænkelig MFC var udviklet. SSMFC har succesfuldt frembragt en maksimal effekt-densitet på 294 mW/m^2 og en tomgangsspænding (OCV) på 1,12 V. Endvidere, blev den reversale spænding undersøgt i detaljer med hensyn til dennes årsag, bestemmelse af parametre og eliminerings metode. Den indre modstand og OCV var de vigtigste parametre for forudsigelse af den reversale spænding. Anvendelse af en kondensator blev fundet til at være en effektiv måde at forhindre reversal spænding og samtidig lagre energi.

En sediment-type MFC baseret på to stykker bioelektroder blev implementeret som en ny *in situ* anvendelses metode til nitrat/nitrit fjernelse, samt el-produktion fra eutrofe søer. Fjernelsen af kvælstof og elproduktion blev begrænset af DO niveauet i vandet og acetat niveauet indsprøjtet i sedimentet. Den foreslåede tilgang kan udvide anvendelsen af sediment MFC-teknologi.

Et nyt nedsænkeligt MDC-system blev udviklet som en *in situ* og ikke-invasiv metode til fjernelse af nitrat fra grundvandet. Systemets ydeevne med hensyn til elproduktion og nitrat eliminerings effektivitet blev undersøgt. Effekterne af hydraulisk retentionstid, ekstern modstand, andre iontyper i grundvandet og forbindelsen med den eksterne nitrifikationsreaktor på systemets ydeevne blev også belyst. Over 90% af nitrat blev fjernet fra grundvandet uden energitilførsel, vandtryk, træk løsning, ekstra elektron donor eller risiko for bakterie udledning. Et sådant nyt system kan tilbyde en spændende mulighed for drikkevands behandling og energiudnyttelse.

Table of contents

List of nomenclature

AEM	Anion exchange membrane
APHA	American Public Health Association
AQDS	9,-10-anthraquinone-2,6-disulfonic acid
ATP	Adenosine-triphosphate
BESs	Bioelectrochemical systems
BOD	Biochemical oxygen demand
CE	Coulombic efficiency
CE_{os}	Overall systemic coulombic efficiency
CEM	Cation exchange membrane
CE_{sub}	Coulombic Efficiency calculated based on substrate consumption
CE_{n}	Coulombic Efficiency calculated based on nitrate removed
COD	Chemical oxygen demand
DGGE	Denaturing gradient gel electrophoresis
DO	Dissolved oxygen
FISH	Fluorescent in situ hybridization
HER	Hydrogen evolution rate
HRT	Hydraulic retention time
MCA	Membrane cathode assemblies
MDC	Microbial desalination cell
MEA	Membrane electrode assemble
MEC	Microbial electrolysis cell
MFC	Microbial fuel cell
OCV	Open circuit voltage
OD	Optical density
OLR	Organic loading rate
ORR	Organic removal rate
PB	Plackett–Burman
PCR	Polymerase chain reaction
PEM	Proton exchange membrane
PFC	Photomicrobial fuel cell
RE_{nG}	Nitrate removal efficiency in groundwater
RE_{nT}	Nitrate removal efficiency of the whole system
RE_{COD}	COD removal efficiency
R_{H2}	Cathodic hydrogen recovery
RVC	Reticulated vitreous carbon
SBMFC	Submersible microbial fuel cell sensor for DO monitoring
SHE	Standard hydrogen electrode
SMDC	Submersible microbial desalination-denitrification cell
SMEC	Self-powered submersible microbial electrolysis cell
SMFC	Submersible MFC
SOM	Sediment organic matter
SND	Simultaneous nitrification and denitrification

SSMFC	Self-stacked submersible MFC
SUMFC	Submersible microbial fuel cell sensor for BOD monitoring
TCV	Total compartment volume
TCD	Thermal conductivity detector
TGV	Total groundwater volume
TNR	Total systemic nitrate removal rate
TSS	Total suspended solid
Y_{H2}	Systemic hydrogen yield

1 Introduction and aim of study

The use of fossil fuels, especially petroleum, in recent years has accelerated a global energy crisis. Furthermore, the combustion of fossil fuels releases more CO_2 to the atmosphere and causes global climate change (2008). Therefore, a carbon-neutral, sustainable energy sources as alternatives to fossil fuels is needed to alleviate the global energy crisis and climate change. A technology using microbial fuel cells (MFCs) in which microorganisms mediate the direct conversion of chemical energy stored in organic matter or bulk biomass into electrical energy has gained considerable interests among academic researchers in recent years (Aelterman et al., 2006b; Chaudhuri and Lovley, 2003; De Schamphelaire et al., 2008b; He et al., 2005; Logan et al., 2006; Min and Angelidaki, 2008; Rabaey et al., 2005a; Thygesen et al., 2009; Zhang et al., 2009c)

MFCs represent a promising technology for sustainable energy production. A typical MFC is consisting of two chambers, i.e. the anaerobic anode chamber and the aerobic cathode. The two chambers are separated by a membrane (e.g., proton exchange membrane) where protons and other ions are transferred from the anode chamber to the cathode chamber, while electrons from the anode chamber are transferred to the cathode chamber through an external electrical circuit and a resistor for electricity production (Booth, 2007; Cheng et al., 2006b; Freguia et al., 2007; Liu et al., 2005b; Liu et al., 2004; Logan et al., 2006; Logan and Regan, 2006a; Min et al., 2008; Zhang et al., 2009b). Various organic compounds and sources of wastes have been successfully utilized for power generation in MFCs (Du et al., 2007; Liu et al., 2005a; Logan et al., 2005; Mathis et al., 2008; Pant et al., 2010; Wang et al., 2008). Additionally, variety of anaerobic bacteria (e.g., *Geobacter* spp.), involved in contaminants reduction, are capable of transferring electrons to a solid electrode and generating electricity in MFCs (Anderson et al., 2003; Bencheikh-Latmani et al., 2005; Tront et al., 2008a; Tront et al., 2008b; Watson and Logan, 2010).

MFCs have been initially developed as a method for simultaneous wastewater treatment and electricity production (Cheng et al., 2006a; Liu et al., 2005a; Logan, 2005; Rabaey et al., 2005a). While interesting, many researchers are realizing that the economic and environmental value of electricity from MFC can not compete that of other energy sources at this stage. Therefore, a development has been initiated recently that expands the scope of MFCs from electricity production to an increasing number of other specialized applications. One

example is MFC-based biosensor which can be applied in different environments for monitoring of organic matter, microbial activity and toxicity (Davila et al., 2011; Kim et al., 2007d; Stein et al., 2011; Tront et al., 2008a). Another successful story is that MFCs can be modified as microbial electrolysis cell (MEC), which have shown promising application for energy rich chemicals production (e.g., hydrogen, methane, H_2O_2) from carbohydrate-rich wastes (Call and Logan, 2008; Cheng and Logan, 2007b; Rozendal et al., 2009; Wagner et al., 2009). MFCs can also be applied in river, lake and marine environments as a method for sediment bioremediation (Donovan et al., 2008; He et al., 2007; Mathis, 2008). Researchers have even invented a new method developed from MFC to reduce the salinity of brackish water or seawater while generating electrical power from organic matters, which is so called microbial desalination cell (MDC) (Cao et al., 2009; Chen et al., 2012a; Mehanna et al., 2010a).

According to above, MFC-based technologies are promising for renewable energy production. However, these technologies are still in their infancy and have not been moved from bench scale operation. These technologies are developed based on MFC, thus they share the common limitations, such as high internal resistance, high energy losses, high costs for construction and operation and difficulty of scaling up (He et al., 2005; Logan et al., 2006; Rabaey et al., 2005b). To overcome some of the present limitations and upgrade MFC-based technologies to field application, efforts are being made to improve the performance and reduce the construction and operation costs.

Therefore, the main objective of this PhD project was to improve the performance, reduce the construction cost, and expand the application scopes of different MFC-based systems. Specific objectives are:

- Development of a novel and cost-effective wastewater treatment process
- Reduction of energy consumption and development of *in situ* applicable MEC reactor for biohydrogen production
- Improvement of sensor design and development of operation strategy for *in situ* and quantitive monitoring
- Expansion of sensor application
- Increase of voltage output and reduction of electrode spacing in sediment MFC
- Expansion of sediment MFC application
- Development of *in situ* applicable MDC for new application

In order to fulfill above objectives, the following work-tasks were addressed.

1 ***Wastewater treatment***

Simultaneous organic carbon, nutrients removal and energy production in a photomicrobial fuel cell (PFC) (*Paper I, Reproduced by permission of the Royal Society of Chemistry*)

To achieve cost-effective and simultaneous C, N, P removal and power generation by MFC, an innovative sediment-type photomicrobial fuel cell (PFC) which is a combination of microalgae cultivation and MFC technologies, was developed. The nutrients removal mechanisms were explored for better understanding the system. Besides, the microbial community enriched in this novel reactor was explored. The effects of algae concentration, COD/N ratio and illumination condition on the PFC performance were detail addressed for further optimization.

2 ***Hydrogen production***

Innovative self-powered submersible microbial electrolysis cell (SMEC) for biohydrogen production from anaerobic reactors (*Paper II*)

To reduce the energy cost and simply the MEC system, a self-powered submersible microbial electrolysis cell (SMEC), in which a specially designed anode chamber and external electricity supply are not needed, was developed. The applicability of the SMEC for *in situ* biohydrogen production from anaerobic reactors was studied. The SMEC performance in terms of hydrogen production, electricity supply, organic matter removal, inhibition of methanogenesis and evolution of microbial community was investigated.

3 ***Biosensor***

Submersible microbial fuel cell sensor for monitoring microbial activity and BOD in groundwater (*Paper III*)

A sensor, based on a submersible microbial fuel cell (SUMFC), was developed for in situ monitoring of microbial activity and biochemical oxygen demand (BOD) in groundwater. The effect of biofilm-colonized or fresh anode on the sensor application in BOD and microbial activity monitoring was investigated. The effect of different operation parameters on the performance of the biosensor was also evaluated. Lastly, the applicability of the biosensor in real contaminated groundwater was verified.

4 *Biosensor*

A simple and rapid method for monitoring dissolved oxygen in water with a submersible microbial fuel cell (SBMFC) (*Paper IV*)

A concept that dissolved oxygen monitoring by means of MFC technology was proposed. A simple and compact reactor design termed submersible microbial fuel cell (SBMFC) was employed to prove the concept. The performance of the SBMFC sensor was evaluated in terms of DO detection range, response time and reproducibility. The effect of different operation parameters (e.g., substrate, temperature, pH, conductivity, nitrate level, external resistance) on the sensor performance was also investigated.

5 *Sediment MFC*

Self-stacked submersible microbial fuel cell (SSMFC) for improved remote power generation from lake sediments (*Paper V*)

To increase the voltage output from lake sediments and shorten the electrodes distance, an innovative self-stacked submersible MFC (SSMFC) was developed. The work was focusing on the voltage reversal occurred in the SSMFC, which is also a major limitation faced by other types of MFCs stack. The cause, determining parameters and elimination of voltage reversal in the SSMFC were studied in detail. The effect of voltage reversal on the microbial community was explored.

6 *Sediment bioremediation*

Bioelectrode-based approach for enhancing nitrate and nitrite removal and electricity generation from eutrophic lakes (*Paper VI*)

To develop an efficient and cost-effective method for nitrate and nitrite removal from eutrophic lakes while expand the application of sediment MFC, a biocathode-based sediment MFC was employed for eutrophic lake bioremediation. The system efficiency was investigated in terms of nitrate/nitrite removal, electricity production and diversity of microbial community. The influence of key factors such as water DO level and organic matter concentration in sediment on the system performance was also explored.

7 *MDC*

A new method for *in situ* nitrate removal from groundwater using submersible microbial desalination-denitrification cell (SMDC) (*Paper VII*)

A new method for in situ nitrate removal from groundwater was developed based on MDC, which has beyond the current application scope of MDC. The performance of the system was investigated in terms of nitrate removal and power production. The removal mechanism was also studied. The effects of operational parameters such as hydraulic retention time, external resistance, and ionic strength on the system performance was investigated.

2 Microbial fuel cell

Microbial fuel cell (MFC) is a bioelectrochemical device that can convert chemical energy stored in waste organic mater or bulk biomass into electricity with the catalysis of microorganisms (Aelterman et al., 2006a; Logan, 2009; Logan et al., 2006; Rozendal et al., 2008a; Zhang et al., 2009c). The electrochemically active microorganisms that responsible for substrate oxidation and electron transfer are the key component of MFC, which makes it different from the traditional chemical fuel cells. In the past decade, MFC has received a great deal of attention as a novel technology for sustainable energy production (Clauwaert et al., 2008; Du et al., 2007; Hamelers et al., 2010; Kim et al., 2007a; Li et al., 2011; Logan, 2010; Logan et al., 2006; Pant et al., 2010; Rabaey and Verstraete, 2005; Schroeder, 2011; Zhou et al., 2011).

The earliest current generation by microorganism was demonstrated by Potter in 1910 (Potter, 1911). However, this finding was not well appreciated until the mid 18th century, when it was discovered that microorganisms could transport the electrons gained from cellular metabolism to insoluble minerals (e.g. manganese) in a process termed extracellular electron transfer (Lovley DR, 1988). A real breakthrough was made when it was found that current and power generation could be enhanced by the addition of electron acceptor. During 1990s, researchers have developed various MFC reactors using domestic or industrial wastewater as substrate which greatly accelerated the progress of technology (Aelterman et al., 2006a; Liu et al., 2004; Logan, 2005; Logan et al., 2006; Rabaey and Verstraete, 2005; Schroeder, 2011). In the last decade, numerous research papers on biological wastewater treatment with MFCs have been published.

The first generation of MFC is driven by electron mediators. In this type of MFC, the microbes are incapable of directly transferring electrons to the anode while external electron acceptors are needed. Electron mediators can easily reduced by accepting the electrons from bacteria and then subsequently release the electrons to the anode and become oxidized status again (Park and Zeikus, 2000). Although mediators can enhance the electron transfer for some bacteria, most of the mediators are toxicity and instable, which limit the applications of mediator-assisting MFCs (Du et al., 2007). The discovery that some microbes can transfer electrons directly to the anode without the help of mediator has lead to the tremendous development of second generation of MFCs, which is called

mediator-less MFCs (Chaudhuri and Lovley, 2003; Kim et al., 1999; Kim et al., 2002). Some of well know electrochemically active bacteria are *Shewanella putrefaciens*, *Geobacteraceae sulferreducens*, *Geobacteraceae netallireducens* and *Rhodoferax ferrireducens* (Bond and Lovley, 2003; Crittenden et al., 2006; Holmes et al., 2004; Kim et al., 2002; Liu et al., 2007b). The finding of these bacteria offers the opportunities to greatly elevate the power production and open the door to intensive MFC studies (Chaudhuri and Lovley, 2003; Schroder et al., 2003).

Comparing traditional bioenergy technologies, the MFC technology has following advantages: (1) broad fuel availability. Nearly all kind of organic matters such as wastewater, sludge and biomass can be utilized as fuel in MFC for electricity production; (2) clean production process and products. A MFC has no substantial intermediary processes, it can convert substrate to electricity directly, which is kind of energy ready for use. There is no second pollution and pollutant production. The off gas is CO_2, which can be discharged without further treatment; (3) less sludge production, due to the electricity production. The bacteria growth yield is considerably low compared to that of anaerobic processes; (4) mild operation condition. Unlike anaerobic digestion and other fermentation processes, MFC can be applied in mild condition such as low temperature and low strength wastewaters; (5) no need of aeration. The air cathode MFC could use oxygen directly from air, thus lower the aeration cost; (6) low cost of catalyst. With the development of bioanode and biocathode, microorganisms could sever as efficient catalyst instead of expensive metals; (7) broad applications. MFC was initially designed for wastewater treatment, but with some modifications, MFC could be easily converted to other kind of technologies for special applications such as pollutant removal, hydrogen production and bioproduction etc (Logan et al., 2006; Rabaey and Verstraete, 2005).

Though promising and has nearly 100 years history (Arends and Verstraete, 2012; Schroeder, 2011), the systematic development of MFC has started only decade ago, MFC is still “young” and has some limitations, such as the performance of the system are far from optimized, the power generation is still low compared with traditional fuel cells; the construction and materials cost are still expensive. However, it should be noticed that the power generation of MFC has increased by several orders of magnitude in the past years. For example, the advances in biocathode research have greatly expanded the application scope of MFCs. It is

reasonably expected that with the development of engineering and science, the MFC and its based technologies have the potential to be more promising renewable energy sources for future.

MFC is not only the foundation but also shares common limitations with other MFC-based technologies, thus a better understanding of MFC technology is important and helpful for optimizing all MFC-based technologies. There, the key aspects of MFC technology are reviewed in this section.

2.1 Working principle

As MFCs are catalyzed by microorganisms, its working principle is obviously different from that of typical chemical fuel cell. The working principle of MFCs can be described with a typical two-chambered MFC, which consists of an anaerobic and an aerobic chamber, physically separated by a proton exchange membrane (PEM) (Figure 1).

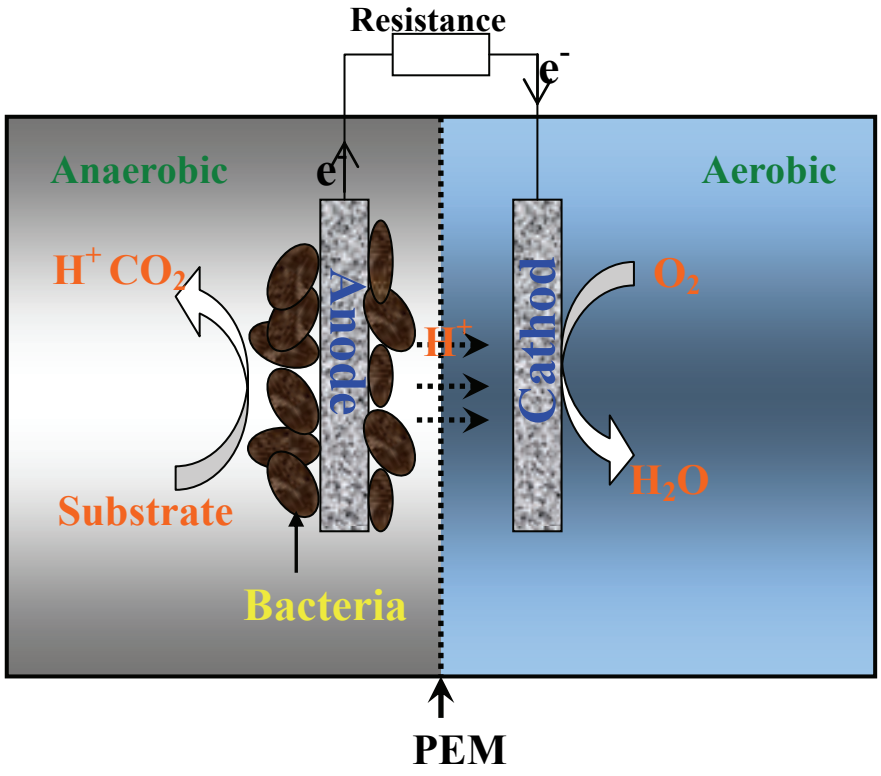

Figure 1. General schematic diagram of a two-chambered MFC

In an MFC, the bacteria attached on the anode electrode oxidize organic substrates and release electrons and protons. Protons in the anode chamber move through the membrane to the cathode chamber, while electrons pass from the

bacteria to the electrode (anode) in the same chamber and then via a circuit to the cathode where they combine with protons and oxygen to form water. In this way, electricity can be produced (Liu et al., 2004; Logan, 2009). Other chemicals such as nitrate, sulfate and manganese could also serve as electron acceptor.

Table 1 Typical electron donors/acceptors related reactions

	Electron donor/acceptor	Reactions
Anode	Acetate	$C_2H_3O_2^- + 4H_2O \rightarrow 2HCO_3^- + 9H^+ + 8e^-$
	Glucose	$C_6H_{12}O_6 + H_2O \rightarrow 6CO_2 + 24e^- + 24H^+$
	Butyrate	$C_4H_8O_2 + 2H_2O \rightarrow 2C_2H_4O_2 + 4H^+ + 4e^-$
	Glycerol	$C_3H_8O_3 + 6H_2O \rightarrow 3HCO_3^- + 17H^+ + 14e^-$
	Malate	$C_4H_5O_5^- + 7H_2O \rightarrow 4H_2CO_3 + 11H^+ + 12e^-$
	Citrate	$C_6H_5O_7^{3-} + 11H_2O \rightarrow 6H_2CO_3 + 15H^+ + 18e^-$
	Sulfur	$HS^- \rightarrow S^0 + H^+ + 2e^-$
Cathode	Oxygen	$O_2 + 4e^- + 4H^+ \rightarrow 2H_2O$
	Bicarbonate[a]	$HCO_3^- + 9H^+ + 8e^- \rightarrow CH_4 + 3H_2O$
	Acetate[a]	$C_2H_3O_2^- + 5H^+ + 4e^- \rightarrow C_2H_6O + H_2O$
	Nitrate	$2NO_3^- + 12H^+ + 10e^- \rightarrow N_2 + 6H_2O$
	Nitrite	$NO_2^- + 2e^- + 2H^+ \rightarrow N_2 + H_2O$
	Permanganate	$MnO_4^- + 4H^+ + 3e^- \rightarrow MnO_2 + 2H_2O$
	Manganese dioxide	$MnO_2 + H^+ + e^- \rightarrow MnOOH_{(S)}$
	Iron	$Fe^{3+} + e^- \rightarrow Fe^{2+}$
	Copper(II)	$4Cu^{2+} + 8e^- \rightarrow 4Cu(s)$
	Potassium Persulfate	$S_2O_8^{2-} + 2e^- \rightarrow 2SO_4^{2-}$
	Ferricyanide	$Fe(CN)_6^{3-} + e^- \rightarrow Fe(CN)_6^{4-}$

[a] External voltage supply is required

The difference in the redox potential between the anaerobic anode (i.e. low redox potential) and aerobic cathode (i.e. high redox potential) is the driven force for electrons transport. Under standard conditions, the maximum potential of the device is ~1.2 V, on the basis of the potential between the electron donor (e.g. NADH) and electron acceptor (e.g. oxygen) (Logan and Regan, 2006b). For example, if glucose is used as substrate and oxygen as electron acceptor, the theoretical maximum potential can be calculated according to the biochemical reactions in MFC shown below:

Anode reaction: $C_6H_{12}O_6 + H_2O \rightarrow 6CO_2 + 24e^- + 24H^+$ (E^o=0.104 vs NHE)

Cathode reaction: $O_2 + 4e^- + 4H^+ \rightarrow 2H_2O$ (E^o=1.229 vs NHE)

A wide range of organic compounds including organic matter in waste/wastewater can be as a fuel of MFCs (Pant et al., 2010). At the cathode side, oxygen is considered to be the most suitable electron acceptor because its high oxidation potential, availability, low cost, and clean end product. In addition, ferricyanide is generally used as electron acceptor, which can increased power density by 1.5 to 1.8 times compared to a Pt covered cathode using dissolved oxygen as electron acceptor (Oh and Logan, 2006). However, toxicity and nonrenewable property of ferricyanide limit its application. There are many specific contaminants (such as nitrate) that can serve as electron acceptors in natural environment (Behera and Ghangrekar, 2009; Clauwaert et al., 2007; Hamelers et al., 2010; Virdis et al., 2008; Paper I; Paper VI). The typical reactions in the anode and cathode of MFCs are listed in Table 1. MFC offers a revolutionary new sustainable energy production technology with simultaneous waste/wastewater treatment.

2.2 Components and materials

A typical MFC consists of three essential physical components which are anode, cathode and separator as shown in Figure 1. In some reactor designs such as single chamber membrane-less MFC, separator (membrane) is leaved out for saving construction cost (Aldrovandi et al., 2009; Liu and Logan, 2004). To achieve power production at a more cost-effective and efficient way, efforts are being made to explore more suitable materials for MFCs.

The selection of both anode and cathode electrodes could follow basic criterions that the material has good conductivity, biocompatible, low resistance, chemically stable and appropriate mechanical strength. There are also some specialized requirement of anode and cathode due to different reactions taken place at these two parts. For example, it is expected that the anode has large surface area which is good for bacteria attaching on (Oh and Logan, 2006). To increase the rate of oxygen reduction at cathode, some electrode modified with catalysts are usually applied (Zhou et al., 2011). The basic component and materials used in MFC studies are summarized in Table 2. The most common anode materials are carbon, available as carbon paper, carbon cloth, carbon mesh, graphite plates, rods, granules or brush, and reticulated vitreous carbon (RVC) (Logan et al., 2006; Zhou et al., 2011). Because of large surface area, high

conductivity and mechanical strength, carbon materials fabricated with nanotechnologies have received significant interest as anode electrode. These electrodes including carbon paper modified with Sn-Pt/MWNT and PPy-CNTs, carbon cloth with CNTs (Qiao and 2007; Sharma et al., 2008; Thepsuparungsikul et al., ; Tsai et al., 2009; Zou et al., 2008). There are also some metal electrodes, including stainless steel, conductive gold and titanium (Dumas et al., 2008; Richter et al., 2008; ter Heijne et al., 2008). In addition of two-dimensional electrode system, three-dimensional electrode such as graphite particles and granular activated carbon has been used to increase the attachment of bacteria and improve the performance of MFCs (Jiang and Li, 2009; You et al., 2008).

Table 2 Basic components and materials of MFCs[a]

Anode	Membrane	Cathode	Cathode catalyst
Graphite rod/ plates/fiber brush	Cation/proton exchange membrane	Graphite rod/ plates/fiber brush	CoTMPP
Carbon paper	Nafion	Carbon paper	Co/Fe/N/CNT
Carbon felt	Ultrex	Carbon felt	PbO_2
Reticulated vitreous carbon	Polyethylene.poly (styrene-co-divinylbenzene)	Reticulated vitreous carbon	β-MnO_2
Carbon mesh	Porcelain	Carbon mesh	FePc
Sn-Pt/MWNT	Anion exchange membrane	Sn-Pt/MWNT	FePcVC
PPy-CNTs	Microfiltration membrane	PPy-CNTs	MnPc
Carbon cloth with CNTs	Bipolar membrane	Carbon cloth with CNTs	Rutile
Stainless steel	Ultrafiltration membranes	Stainless steel	Co-OMS-2
Gold	J-Cloth	Gold	MnO_x
Titanium	Glass fibers	Titanium	Fe^{3+}
	Porous fabrics	Aluminum foil	Polyaniline

[a] The references for each material are given in the corresponding text.

Generally, the materials used as anode can also be applied as cathode. The common cathode materials are carbon paper, carbon cloth, carbon brush, graphite (Zhou et al., 2011). In order to increase power generation, various highly active catalysts have been used to modify cathode. Pt is the most popular catalyst, which could reduce the cathodic reaction activation energy and increase reaction rate at cathode (Yang et al., 2009; Paper I; Paper II; Paper III; Paper V). However, Pt is too expensive to practical application. More efforts are making to reduce the

loading of Pt or searching for other cheap alternative catalyst, including CoTMPP, Co/Fe/N/CNT, PbO_2, β-MnO_2, FePc, FePcVC, MnPc, Rutile, Co-OMS-2, MnO_x (Cheng et al., 2006b; Deng et al., 2010; Lu et al., 2011; Morris et al., 2007; Zhang et al., 2009a; Zhao et al., 2006; Zhou et al., 2011).

Except sediment MFC and single chamber membrane-less MFCs (Donovan et al., 2008; Wang et al., 2008), most of MFC designs require the separation of the anode and cathode chamber using a membrane. The earliest separator is salt bridge, but due to its high internal resistant, it is seldom used in recent MFC studies (Min et al., 2005). Currently, cation exchange membranes (CEM), which are also referred to proton exchange membranes, have been widely used (Kim et al., 2007b). Several non-expensive separator have also been developed, including anion exchange membrane (AEM), bipolar membrane, microfiltration membrane, ultrafiltration membranes, porous fabrics, glass fibers, J-Cloth (Kim et al., 2007b; Li et al., 2011).

Generally, anode, cathode and membrane are separated in MFCs. Membrane electrode assemble (MEA) has recently emerged as an efficient separator-electrode configuration to decrease internal resistance and improve the power generation of MFCs (Paper II and paper V). MEAs are divided into two different categories: membrane cathode assemblies (MCA) and anode-membrane-cathode assemble (sandwich-type) MEAs (Li et al., 2011). The construction of these two different types of MEA is shown in Figure 2. The use of MCA by hot-pressing a CEM directly onto a carbon cloth cathode has led to the invention of first single-chamber air cathode MFC, which greatly simplified reactor design and reduced construction and operation cost of MFCs (Liu and Logan, 2004). To assemble anode, membrane and cathode together by hot-pressing has greatly minimized electrode spacing and thus reduced the ohmic resistance of MFCs (Kim et al., 2009). A variety of cost-effective materials have been used to construct MEAs including membrane, graphite paint, J-cloth, canvas cloth, cellulose dialysis tube and polycarbonate filter (Biffinger et al., 2007; Kim et al., 2007a; Li et al., 2009; Li et al., 2011; Rabaey et al., 2005b; Zhuang et al., 2009; Zuo et al., 2008). The invention of MEAs has opened up a new avenue toward MFC operation, scaling-up and application.

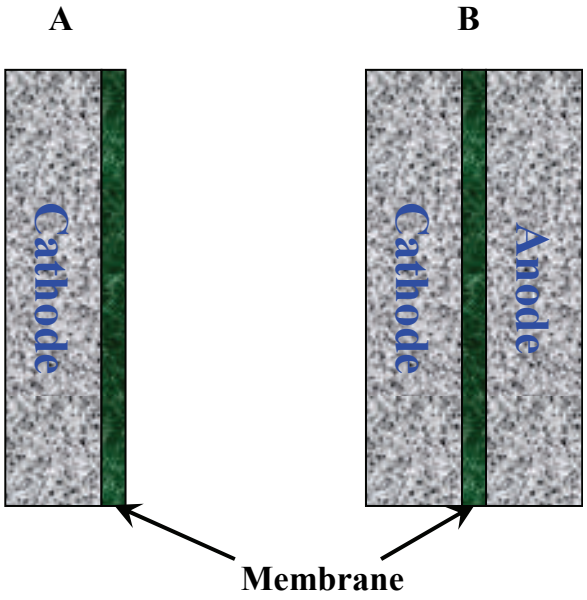

Figure 2. Schematic of MEA configuration: (A) membrane cathode assembly; (B) sandwich-type MEA.

2.3 Reactor design

System architecture is important for the performance of MFCs. It has been recognized that improvement of the reactor design could significantly contribute to overcome some of the present limitations. With more than ten years intensive research, many different configurations of MFCs ranging from 1.5 μL to several liters have been developed. Generally, according to the numbers of reactor chamber, MFCs could be clarified as two-chamber, single-chamber and multi-chamber i.e. stack systems.

2.3.1 Two-chamber MFC systems

Two-chamber MFCs are commonly used for lab-based batch test especially in earlier studies. "Two-chamber" refers to the anode and cathode chambers separated by a separator (e.g., membrane), which allows protons to transfer across to the cathode while prevent the diffusion of cathodic electrolyte to the anode. The construction principle of two-chamber MFCs is shown in Figure 1. Two-chamber MFCs can be further classified according to the different shapes of the two chambers (Du et al., 2007). The typical systems are salt bridge MFC, cylindrical MFC, rectangular and flat MFC, miniature MFC and up-flow tubular MFC, as summarized in Table 3.

Salt bridge is an inexpensive material made of glass tube that is heated and bent

into a U-shape and filled with agar and salt, which could serve the same function as a proton exchange membrane. The salt bridge MFC is typically constructed by linking two glass bottles with salt bridge. Min et al (2005) constructed a salt bridge MFC by joining two media bottles with a U-shaped glass tube salt bridge filled with PBS. Though the salt bridge is much cheaper than a proton exchange membrane, little power can be produced from this type of MFC due to its very high resistance, which limits its applications.

"H" shape MFC is a widely used two-chamber MFC design, consisting usually of two glass bottles or cylindrical chambers connected by a membrane, which is usually CEM such as Nafion etc descried in Section 2.2. H-type MFCs are operated in batch mode and suitable for basic parameter research such as examining power generation from specific substrate or testing the performance of new materials, or exploring the microbial communities enriched from different substrates, but power generation is relatively low in such system. The power generation of these systems is limited by high internal resistance, small electrode surface area and membrane selectivity (Logan et al., 2006).

Rectangular or flat MFCs have been developed to reduce the electrode spacing and enlarge electrode surface. Rabaey et al. (2003) developed a rectangular MFC where the chambers are separated by a membrane. In such configuration design, four cells are connected into one block, thus giving rise to four cathodes and anodes. A combined electrode/proton exchange membrane system was used in a flat plate MFC to further shorten the electrode distance (Min and Logan, 2004). The flat plate MFC was designed to operate as a plug flow reactor. The reactor consisted of two chambers which were separated by the electrode/membrane assembly (see Section 2.2). The special design of flat plate or rectangular MFC is benefit to shorten the electrode spacing and increase electrode surface, but large area of membrane is also required which may increase the construction cost.

Miniature MFC have received increasing interests as they can provide unique platforms for fundamental studies of microbes such as screening environmental strains and as a portable power source for small electronic elements (Qian et al., 2011). A 1.2 mL MFC that demonstrates high output power (24 and 10 mW/m^2 based on the true surface areas of reticulated vitreous carbon (RVC) and graphite felt) has been developed by Ringeisen et al (2006). A 650 μL MFC array contains 24-well was used to isolate an electrochemically active microbe that produces 2.3-fold higher power than the wild-type *Shewanella oneidensis*

MR-1(Hou et al., 2009). A MFC equipped with a nanoelectrode provide a new approach to investigate extracellular electron transfer at the single-cell level (Jiang et al., 2010). Qian et al. (2011) demonstrated an easily fabricated, PDMS-based, sub-5 μL MFC that generates an enhanced power density of 62.5 W/m^3. Miniature MFCs are also facing some limitations such as low power densities and high fabrication costs (Qian et al., 2011).

Up-flow mode MFCs are easily to scale up and thus more suitable for wastewater treatment, compared to previous two-chamber reactors. He et al. (2005) designed an upflow MFC which was fed with artificial wastewater and operated in continuous mode. The internal resistance of this system is 84 Ω which limits the power output. In order to further reduce the internal resistance, the same researchers developed an upflow MFC system with a U-shaped cathode inside the anode chamber, which has a small internal resistance of 4 Ω. A maximum volumetric power of 29.2 W/m^3 was produced at a loading rate of 3.40 kg COD/m^3/day and an operating temperature of 35 °C while feeding sucrose continuously (He et al., 2006). Normally, fluid recirculation is required in the up-flow mode MFCs, which may increase the operation costs and minimize the benefit of microbial electricity production (Du et al., 2007).

Sediment MFC is kind of two-chamber system. Electricity can be harvested by embedding an anode into the sediment, and connecting it through an electronic circuit to a cathode in the overlying oxygenated water (Bond et al., 2002; Paper V; paper VI). Though there is no real separator for separating the anode and cathode, the solid and water phases can be a naturally anode chamber and cathode chamber, respectively. This system can be employed for sediment bioremediation or as power sources for fresh water or marine studies (Donovan et al., 2008; He et al., 2009a; Holmes et al., 2004). The main limitations of sediment-type MFC are their low power and output voltage. Several attempts to increase the power output have been made, including modifications of the electrode material, alterations of electrode design, adding mediators or particulate substrate to the anode (Hasvold et al., 1997; He et al., 2007; Hong et al., 2009; Rezaei et al., 2007; Paper V).

Table 3 Summary of different reactor designs[a]

Chambers	Type of reactor	Advantages	Limitations
Two	Salt bridge	Cheaper materials	High internal resistance
Two	H-shape	Basic research/stable	Large electrode spacing/ low power density
Two	Rectangular/ flat	Short electrode distance	Expensive membrane
Two	Miniature	Portable power source	low power, high fabrication costs
Two	Up-flow tubular	Scalable, continuous operation	Expensive membrane/ fluid recirculation
Two	Sediment MFC	Simple	Low power generation
Single	Air cathode with membrane	Simple and compact	Expensive membrane
Single	Air cathode without membrane	Simple, compact and scalable	Low CE
Single	Up-flow tubular	Scalable, continuous operation	Low CE/toxicity ferricyanide
Single	Up-flow rectangular	Fully mixing/ DO gradient	Low CE/H_2O_2 consumption
Single	Up-flow cylindrical	Glass wool/bead as separator	High internal resistance
Single	Submersible MFC	Simple, *in situ* applicable	Aeration/ high internal resistance
Twelve	6 unites stack MFC	High power and voltage output	Voltage reversal
Two	two-cell air-cathode stack	Foundation study	Voltage reversal
Four	CEM bridged stack	High cations transfer/low internal resistance	Expensive membrane
Five	Bipolar plate stack	Minimized voltage drop	Voltage reversal

[a] the references for each design are given in the corresponding text.

2.3.2 Single-chamber MFC systems

As mentioned earlier, two-chamber systems are suitable for foundation studies, but it is difficult to scale-up them for practical application. Thus, more simple and compact configurations such as one-chamber system are required for reducing construction and operation cost. One-chamber MFCs typically possess

only an anode chamber without aeration in a cathode chamber. In the typical one-chamber MFC, the anode and cathode electrodes are placed on either side of a tube. A flat plate is put against the anode and one side of the cathode is exposed to air directly while the other side faces to water (Liu et al., 2005b). The advantages and disadvantages of typical single-chamber systems are summarized in Table 3.

Membrane is initially used in single chamber, air cathode MFCs to keep the water from leaking through the cathode and prevent oxygen diffusion into the anode (Liu et al., 2005b). To reduce the construction cost, Liu and Logan (2004) further developed a reactor called single chamber, air cathode membrane-less MFC. The power generation from this system is much higher than that of single chamber, air cathode MFC. However, Coulombic efficiency (CE) was only 9-12% without membrane and 40-55% with the PEM, indicating substantial oxygen diffusion from the cathode into the anode chamber in the absence of the PEM. Some cheap materials such as Polytetrafloroethylene (PTFE) are used to support the outside of cathode to prevent bulk water loss because of hydrostatic pressure on the cathode (Cheng et al., 2006b).

Several variations on single-chamber MFC design have emerged in the effort to pursue a scalable reactor design or improve the power generation. Park and Zeikus (2003) developed a single chamber MFC composing of a rectangular anode chamber with a porous cathode exposed to the air directly. Rabaey et al. (2005b) constructed a tubular, single-chambered, continuous MFC that produces high power using a granular graphite matrix as the anode and a ferricyanide solution as the cathode. Jang et al. (2004) developed a membrane-less MFC with cylindrical shape, which is also operated in continuous flow mode. The reactor made of a Plexiglas cylinder was separated into two sections by glass wool and glass bead layer. The separator used in this system is cheaper than PEM. Tartakovsky and Guiot (2006) designed a similar reactor but in rectangular and without using any separator. Min and Angelidaki (2008) developed a submersible MFC (SMFC) by immersing an anode electrode and a cathode chamber in an anaerobic reactor. The advantage of such reactor design is that no special anaerobic anode chamber is needed, as the cathode chamber and anode electrode could be immersed in existing anaerobic reactors or natural anaerobic environments (Min and Angelidaki, 2008; Paper III). The single-chamber MFCs have greatly reduced the construction cost and accelerated the application of MFC-based technologies, which offers new opportunities to reactor design for

future studies.

2.3.3 Multi-chamber/stacked MFC systems

To increase the overall system voltage or current, MFCs can be connected in series or parallel as a stacked system. Connecting several MFCs in parallel adds the current, while one common voltage applied to each cell. In case several MFCs are connected in series, the voltage is added. Therefore, any desired current or voltage could be obtained by parallel and series stacking the appropriate number of MFCs (Aelterman et al., 2006b; Paper V).

Several reactor designs have been developed for this purpose. The first MFC stack is developed by Aelterman et al. (2006b). Their MFC stack consists of six individual units with granular graphite anode. The MFC stack produced a voltage of 2.02 V at 228 W/m^3 while current of 255 mA are produced at 248 W/m^3 in parallel connection. Both open circuit voltage and short circuit current were approximately a factor of 6 higher than that of the individual MFCs. However, voltage reversal occurred in some cells at high power density during series connection, which resulted in a rapidly decrease of power and voltage output. The authors owed it to the lacing of microbial activity. In order to disclose the cause of voltage reversal in stacked MFCs, Oh and Logan (2007) constructed a two-cell air-cathode MFC stack which produced a working voltage of 0.9V at 500 Ω and an open circuit voltage (OCV) of 1.3V when operated in fed batch mode with sufficient substrate supply. The authors found that the voltage reversal is due to loss of bacterial activity at the condition of fuel starvation. A novel configuration of stacked MFCs bridged internally through an extra CEM was assembled from two single MFCs (Liu et al., 2008). To minimize the limitations that non-uniform potential distribution on the electrode surface and lower output voltage due to the potential drop, a bipolar plate stacked MFC consisting of five single cells connected in series was developed (Shin et al., 2006). In Paper V, a novel stack configuration was developed, which can produce high voltage from lake sediments, a strategy for eliminating voltage reversal was also proposed.

2.4 Key parameters describing the performance

Due to the variation of reactor design and operation conditions adopted by researchers, uniform of data reporting is required to compare the results among different systems. Table 4 summarizes the main parameters that have been used to evaluating the performance of MFCs.

Table 4 Parameters for evaluating MFC performance

Parameter	Unit	Calculation/measurement[d]
Electrode potential	V	$E=E^o-RT/(nF)\ln(a_{red}/a_{oxy})$[a]
OCV	V	Voltage at indefinite resistance
Current	A	$I=E/R$, E is voltage, R is external resistance (Ω)
Power	W	$P=E^2/R$ or $P=IE$
Current density	A/m^2	$I_A= I/A$, A is projected electrode surface area (m^2)
Power density (surface area)	W/m^2	$P_A=E^2/R/A$
Volumetric power density	W/m^3	$P_A=E^2/R/v$, v is the reactor volume (m^3)
Coulombic efficiency[b]	%	$CE=M\int_0^{t_b} Idt \Big/ Fbv_{An}\Delta COD$
Energy efficiency[c]	%	$\varepsilon = \int_0^t EIdt \Big/ \Delta H m_{sub}$
Internal resistance	Ω	$P_{\max} = OCV^2 R/(Ri+R)^2$, calculated from slope of the polarization curve, R_i internal resistance (Ω)
Treatment efficiency	%	$\eta = \Delta C/C * 100\%$, ΔC is removed substrate (kg), C is total substrate fed (kg)
Organic loading rate	Kg/m^3/day	$OLR = C_i Q / v_{An}$, Ci is substrate concentration (kg/m^3), Q is feed flowrate m^3/day
Organic removal rate	Kg/m^3/day	$ORR = \Delta C/(v_{An} t_b)$
Hydraulic retention time	day	$\mathrm{HRT} = v_{An}/\mathrm{Q}$

[a] E^o is the standard potential; R is the gas constant; n number of electrons involved in the reaction; F is Faraday constant a_{red}/a_{oxy} is the activity ratio of reductant and oxidant

[b] M is the molecular weight of oxygen; t_b is the reaction time; n is the number of electrons exchanged per mole of oxygen; v_{An} is the liquid volume in the anode

[c] ΔH is the heat of combustion (J/mol) and m_{sub} is the amount (mol) of substrate.

[d] The references for each calculation are given in the corresponding text.

The potential between the anode and cathode is an important parameters determining the power generation as it is the driving force for electron moving. The theoretical maximum voltage can be generated by an MFC is dependent on

the electrode potentials of anode and cathode. Thus, the calculation should be made according to the electrochemical reactions that occur between the anode electron donor (i.e. substrate) at a low potential and the cathode electron acceptor with a high potential (Du et al., 2007). The electrode potential can be calculated by Nernst equation as shown in Table 4. The real electrode potential can only be measured with a reference electrode with a known potential. The common reference electrodes used are silver-silver chloride Ag/AgCl standard hydrogen electrode (SHE) or normal hydrogen electrode (NHE) (Logan et al., 2006). OCV is the maximum voltage can be really produced by MFCs, which is lower than the maximum theoretical voltage. OCV can be determined by connecting MFCs with an indefinite resistance or measuring the voltage between anode and cathode in the absence of current (Watanabe, 2008).

In most of case, MFCs are connected with an external resistance, while the voltage loss (i.e. working voltage) on the external resistance is monitored with commonly available voltage meters, multimeters and data acquisition systems. The measured voltage is used to calculate electric current according to Ohm's law. Power, calculated based on the current and voltage, is the mostly used parameter for reporting the MFCs performance. To compare the power output from different systems, power and current is often normalized to electrode surface area (Logan et al., 2006). In case for engineering calculation of reactor size and construction cost, power is normalized to the reactor volume (Table 4).

Coulombic efficiency (CE) is calculated as the ratio of total amount of Coulombs actually produced and to the total Coulombs contained in substrates. The actually transferred Coulombs can be determined by integrating the current over time (Table 4) (Logan et al., 2006). Energy efficiency (ε) is another important parameter for evaluating how efficient of MFC compared with more traditional techniques such as anaerobic digestion. The systematic energy efficiency is calculated as the ratio of power actually produced by MFC to the heat energy obtained by substrate combustion (Logan et al., 2006; Watanabe, 2008). Polarization curve is a powerful tool for calculating the internal resistance, OCV and maximum power density in MFCs studies (Logan et al., 2006). Polarization curve can be made by measuring current at different voltage using a potentiostat or measuring voltage at different external resistance (Watanabe, 2008). Internal resistance is equivalent to the slope of the polarization curve. In continuous operation, organic loading rate (OLR) is useful to evaluate the treatment capacity of different MFC systems, which can also be used to compare MFCs with

traditional treatment technologies. Corresponding to OLR, organic removal rate (ORR) is used to evaluate the treatment capacity of MFCs. MFCs have initially proposed as a wastewater treatment technology, and thus parameters used in traditional treatment technologies are also applicable to MFCs, such as biological oxygen demand (BOD), chemical oxygen demand (COD), or total organic carbon removal efficiency (Logan et al., 2006). Hydraulic retention time (HRT) is a parameter describing the average time the wastewater remains in MFCs.

2.5 Factors affecting the performance

To improve the performance and reduce the construction, operation and maintenance cost of MFCs, affecting factors need to be clearly addressed. These factors can be classified into three catalogues: system architectures, operational conditions and biological factors.

2.5.1 Architectural factors

Generally, reactor design and related materials selection is important for performance and updating system to large scale application. The main architectural factors are reactor type, electrode spacing, presence, type and size of separator, ratio of electrode surface area and volume, electrode materials and catalyst selection, liquid or aerated cathode (Borole et al., 2011).

Reactor design affecting both the performance and operation and construction cost of MFCs. In section 2.3, numerous reactor configurations have been designed. Tow-chamber systems are normally used for basic or foundation investigation, as this kind of reactor have high internal resistance due to large electrode spacing. In addition, the two chambers design will add the construction cost which is not suitable for field application (Clauwaert et al., 2008). In this background, single-chambers air cathode MFC seems more attractive for practical application.

Separator utilization especially use of various membranes can affect the internal resistance and construction cost of MFCs, but it is helpful for minimizing the coulombic losses (Liu and Logan, 2004). A CEM has slow proton transfer capacity and could result in a rapid accumulation of acidity in the anode, which can decrease the activity of exoelectrogens (Harnisch et al., 2008). Introducing anode effluent to the cathode has been proposed to alleviate the imbalance of pH (Freguia et al., 2008). Omitting membrane from MFC can also be an effective

way to balance pH in the anode and cathode (Liu and Logan, 2004).

Electrode properties can affect both the aggregation of exoelectrogenic biofilm and electron transfer in MFCs. The electrode materials can affect biofilm formation, structure and microbial communities. Different materials also contribute differently in internal resistance. It has been reported that use of electrode materials with high friendly microbial-accessible surface (such as carbon fiber or paper) improved 40% of current generation (Liu et al., 2010). Electrode and anode chamber design (e.g., ratio of electrode surface area and volume) can significantly affect the protons and substrate transportation (Borole et al., 2011). Using nanotechnologies to modify the surface of electrode or application of effective electrode catalysts may help maximum the surface area and enhance the activity of electrode (Deng et al., 2010).

2.5.2 Operating factors

Beside reactor design, the performance and microbiology of MFCs is influenced by the operational parameters applied, such as external resistance, substrate type, concentration and feeding rate, pH, temperature, conductivity/ionic strength, mixing velocity/shear rate.

External resistance is an important electrical factor for power generation, which controls the ratio between the electric current and the working voltage (Borole et al., 2011; Paper V). A low external resistance leads to a low working voltage and high current, which results in a high substrate conversion rate; the opposite is true in the case of a high external resistance. In principle, the maximum power can only be obtained when the external resistance equal to the internal resistance of fuel cells. Thus, by changing the external resistance and recording the voltage and current produced, the internal resistance can be estimated (Logan et al., 2006). It has been reported that the external resistance can affect the microbial composition of anode biofilm (Lyon et al., 2010). Using an external resistance equal to or lower than the internal resistance has been proposed to promote biofilm growth and maximize the power generation of MFCs (Aelterman et al., 2008). In Paper V, we found that the external resistance can affect the performance of a MFC stack and low external resistance contribute to voltage reversal

pH, temperature, conductivity/ionic strength and shear rate are typical parameters both in batch and continuous operation (Paper III and Paper IV). pH

and temperature are two of the most important environmental factors impacting bacterial cell growth and physiology. The effect of pH on power generation has been well addressed. The pH gradients formed in MFCs by using CEM can disturb the operation of MFCs and decrease their performance and durability (Biffinger et al., 2008). A neutral pH in anode while high acidity in cathode is preferable for electricity production (Borole et al., 2011). Temperature affect not only bacteria growth but also the power generation of MFCs (Patil et al., 2010b). Normally, power generation is increase with environmental temperature. It has been observed that increasing temperature from 30 to 40 ^{o}C increased current generation by 80% (Liu et al., 2010). Min et al. (2008) found that a lag phase of 30 h occurred at 30 ^{o}C which was half that at 22 ^{o}C. The maximum power density was 70 mW/m^2 at 30 ^{o}C while 43 mW/m^2 was produced at 22 ^{o}C. They also observed 4 times higher power density with phosphate buffer addition (conductivity of 11.8 mS/cm) than the value without phosphate additions (2.89 mS/cm). It has been reported that high shear rates can enhance aggregation and attachment of microbes and thus more compact biofilms in many microbial systems (Pham et al., 2008). Pham et al.(2008) observed that biofilms formed at shear rate of 120/s were twice as thick and 5 times biomass density than those at shear rate of 0.3/s. The microbial community composition was also significant different. The current density was 2-3 times higher at higher shear rate than that at low shear rate. 30% increase of current density was observed when increasing the liquid circulation from a Reynolds number of 900 to 4900 (Borole et al., 2011).

Substrate type and Loading rate are important factors determining the character and amount of substrate fed into MFC reactors. It has been reported that the microbial community and power generation were different when different type of substrates (fermentative and non-fermentative) were fed as fuel in MFCs (Jung and Regan, 2007; Kim et al., 2007c; Zhang et al., 2010). A step-wise increase of the loading rate, either by changing the substrate concentration or the flow rate, can successively lead to an increase, saturation and decrease of the power generation during polarization (Borole et al., 2011; Moon et al., 2006; Rabaey et al., 2003).

2.5.3 Biological factors

The most important biological factors affecting the performance of MFCs are the type and source of inoculums. The inoculum determines the growth of biofilm,

electron transfer mechanism and rates, biofilm thickness and conductance and substrate uptake rate etc, which could further affect the activity of biofilm and electricity production (Borole et al., 2011). Pure cultures such as *Geobacter* and *Shewanella* are suitable for foundation studies and have high electrochemically activity, while mixed cultures are more suitable for practical application since the conditions for pure culture is difficult to keep (Bond and Lovley, 2003; Logan et al., 2006). Gram-negative microorganisms normally can produce higher power than Gram-positive bacteria because the different cell structures of these two types microorganisms (with or without outer membrane) (Borole et al., 2011). Inoculums used in MFCs are generally taken from various wastewaters, sludge, sediments, soil (Bond et al., 2002; De Schamphelaire et al., 2008b; Ki et al., 2008; Liu et al., 2007a). Beside exoelectrogens, there are also competitive microorganisms existing in the anode of MFCs such as denitrifying bacteria, hydrogen-scavenging microorganisms and methanogens. These microorganisms can compete with electrochemically active bacteria and consume electrons, which results in energy losses and lower the power generation (Borole et al., 2011; Ishii et al., 2008). Thus, minimizing the growth of non-electrochemically active microorganisms is important to the development of MFCs. In Paper V, we have adopted an effective strategy for inhibiting the growth of methanogens.

3 MFC-based technologies and their applications

MFC is capable of converting the chemical energy stored in the organic matters to electricity assisted by microorganisms and thus is considered as environmental friendly conversion technology. MFCs are initially emerged as an alternative energy-added wastewater treatment technology which can produce electricity during wastewater treatment. At the earliest stage of development, researchers expected that the energy gained from wastewater treatment could at least cover the cost for wastewater treatment and this technology indeed shows a promising perspective for this purpose.

However, as a new technology, there are still many limitations need to be solved before moving to field application. For example, even the power generation of MFCs has been improved by several orders of magnitude in the past decade, the maximum power generations at present are in the order of 10-100 W/m^3 MFC, which is still far from the value (400 W/m^3) at which MFC can compete to anaerobic digestion (Clauwaert et al., 2008; Pham et al., 2006). There are several limitation or affecting parameters need to be optimized, as described in section 2.5. Therefore, researcher is realizing that before MFCs become more competitive energy production and conversion technologies, the application field of MFCs should have to be shifted from electricity production toward to specialized value-added applications.

Considering above, several MFC-based technologies have been developed for various applications such as wastewater and specific pollutant treatment, biosensor, sediment bioremediation and as remote power sources (Aelterman et al., 2006a; Clauwaert et al., 2008; Logan et al., 2006; Paper I; Paper III; Paper IV; Paper V; Paper VI). The discovery that electrical current can drive microbial metabolism has recently resulted in several new applications, such as MEC for hydrogen, methane, ethanol production (Call and Logan, 2008; Steinbusch et al., 2010; Wagner et al., 2009; Paper II). Furthermore, a new method developed from MFC to reduce the salinity of brackish water or seawater while generating electrical power from organic matters has draw much attention, which is so called microbial desalination cell (MDC) technology (Cao et al., 2009; Chen et al., 2012a; Mehanna et al., 2010a; Paper VII). Notably, the microbial assisted chemicals production, called microbial electrosynthesis, offers a highly attractive,

novel route for producing valuable products from wastewater with small amount electricity added (Rabaey and Rozendal, 2010). The knowledge gained from successful near-term applications will improve the energy production of MFCs-based technologies which is a long-term prospect.

3.1 Wastewater treatment

The possibility that MFCs can be used for wastewater treatment was first proposed in 1911 (Habermann and Pommer, 1991). Wastewater treatment based on MFCs offers a great opportunity to develop the technology, as the fuel for electricity production is free "wastewater" which is must be treated. An MFC would replace the existing energy-coasting bioreactors (e.g., activated sludge system), resulting in an energy-producing system which can recover energy from wastewater (3.8 kWh/Kg COD) without aeration cost (1KWh/kg COD in conventional aeration). Moreover, MFC process has less sludge production of 0.02-0.22 g biomass-COD/g substrate-COD compared with 0.53 g biomass-COD/g substrate-COD for conventional aerobic treatment (Clauwaert et al., 2008).

3.1.1 The removal of organic matters

Until now, various artificial and real wastewater have been treated with MFCs, such as brewery wastewater, beer brewery wastewater, chocolate industry wastewater, domestic wastewater, food processing wastewater, meat processing wastewater, paper recycling wastewater, protein-rich wastewater, real urban wastewater, starch processing wastewater and swine wastewater (Pant et al., 2010). The organic carbon removal from the anode of MFCs is the first strategy, inorganic matters removal in the anode is another. For example, sulfide and ammonia oxidation have been successfully performed in the anode of MFCs (He et al., 2009b; Rabaey et al., 2006).

3.1.2 Shift to specific pollutants treatment

While microbial oxidation at the anode may be primarily used for organic matters removal, the discovery of biocathode and related reduction processes at the cathode provide an opportunity to expand the application of MFCs in wastewater treatment. In this context, several pollutants such as nitrate, nitrite, perchlorate, chlorinated compounds, copper, mercury and iron can be removed (Aulenta et al., 2007; Clauwaert et al., 2007; Tao et al., 2011; Ter Heijne et al., 2007; Thrash et

al., 2007; Virdis et al., 2008; Wang et al., 2011; Paper I; Paper VI).

One typical application is nitrogen species removal in MFCs (Paper I and Paper VI). The first study of nitrate denitrification in MFC was demonstrated by Clauwaert et al. (2007), where a complete denitrification at cathode without extra electron donor supply was successfully performed in a tubular reactor. At the same period, Lefebvre et al. (2008) investigated the same process in a two-chambered MFC. In order to further improve this application, Virdis et al. (2008) demonstrated a novel process which integrates MFC with aerobic nitrification for simultaneously carbon and nitrogen removal. In their "loop configuration" system, the effluent from the anode of a synthetic wastewater-powered MFC was fed to an external aerobic nitrification vessel for oxidation of ammonium to nitrate. This nitrate-enriched stream was subsequently fed back to the MFC cathode for denitrification. To further solve the problem that ammonia diffusion from anode to the cathode through CEM, Virdis et al. (2010) integrated the nitrification stage into the cathode chamber, where simultaneous nitrification and denitrification (SND) were accomplished. Other systems for the same purpose have been developed. A combined use of the membrane aerated biofilm and MFC processes was proposed (Yu, 2011). Xie et al. (2011) developed an oxic/anoxic biocathode system for simultaneous carbon and nitrogen removal. A simultaneous nitrification and denitrification was also achieved in a single-chamber MFC pre-enriched with a nitrifying biofilm (Yan et al., 2012).

Another approach to produce more added values from MFCs is the combination of MFCs with existing wastewater treatment technologies. Integration of MFCs with anaerobic digestion has been proposed to remove sulfides and residual organic matters (mainly of volatile fatty acids) from digester effluents (Aelterman et al., 2006a; Pham et al., 2006; Rabaey et al., 2006).

3.2 Biosensors

Another profitable application of MFCs is as biosensor for environmental monitoring.

3.2.1 BOD monitoring

BOD monitoring is one of typical applications of MFC-type biosensor, since the current generation is dependent on the microbial respiration and proportional to

the fuel concentration (Kim et al., 2003; Paper III). Several types of MFC-based biosensor have been developed for monitoring of BOD in surface water, secondary effluent or wastewater samples and showed good stability, accuracy and wide detection range when compared with other types of biosensors (Chang et al., 2004; Di Lorenzo et al., 2009a; Di Lorenzo et al., 2009b; Kang et al., 2003; Kim et al., 2003). The linear relationship between Coulombic yield of MFCs and strength of the wastewater has been firstly employed for BOD monitoring. Kim et al. (2003) tested this concept with a two-chamber MFC. Their sensor gave a good correlation between the BOD value and the coulomb produced and it has been operated for over 5 years in a stable manner without any servicing. Later, researchers realized that electric current is more suitable as an indicator than Coulombic yield for BOD monitoring, as Coulombic yield is calculated only after the BOD has been consumed and thus a high BOD level requires a long response time (Du et al., 2007). Efforts have been made to improve the dynamic responses of MFC sensor. Moon et al. (2004) investigated the dynamic behavior of a continuous mediator-less MFC as BOD sensor, which had the shortest response time of 36 min. Various types of MFC reactors including two-chamber and single chamber systems have been used as BOD sensor and BOD levels from 2 to 350 mg/L can be detected in wastewater (Di Lorenzo et al., 2009b; Moon et al., 2005). To develop a MFC type BOD sensor having enhanced performance and providing high sensitivity and accuracy, membrane-electrode assembly has been employed (Kim et al., 2009).

3.2.2 Microbial activity assessment

Microbial activity monitoring has been recently proposed as another possible application of MFC sensor (Tront et al., 2008a; Tront et al., 2008b; Williams et al., 2010b; Paper III). The idea behind of this application is that the response of current generation to different substrate (BOD) concentrations somehow reflects the activity of the biofilm colonized on the anode (Paper III). Williams et al. (2010b) developed an electrode-based approach by placing anode electrode in anoxic subsurface environments for monitoring of microbial activity. In such system, *Geobacter* species in the subsurface environments readily colonized the graphite electrode and produced current which correlated to the availability of acetate added to promote U(VI) reduction. This method could be used to detect microbial activity in other planet. Abrevaya et al.(2010) reported a cylindrical reactor without anode chamber for extraterrestrial microbial life detection.

3.2.3 Chemical toxicants monitoring

Toxicity monitoring is recently proposed based on the principle that the inhibition of electrical current can be a good indicator for the appearance of toxicants, which has expanded the application scope of MFC sensors (Kim et al., 2007d). Chemical toxicants can inhibit the metabolic activity of electrochemically active microorganisms, which can subsequently inhibit the transfer rate of electrons to the electrode and thus decrease the electricity production. Therefore, MFCs has been proposed as a toxicity sensor for monitoring the presence of chemical toxicants in various environments (Stein et al., 2012).

Patil et al. (2010a) investigated the effect of exemplary biocides on wastewater-derived electroactive microbial biofilms and found that the current generation of a mediator-based MFC enriched with planktonic cells from wastewater was massively affected by the presence of the antimicrobial agents. To obtain a simple, compact and planar device for toxicity monitoring, Dávila et al. (2011) developed a novel silicon-based MFC. Their device consists on a proton exchange membrane placed between two microfabricated silicon plates that act as current collectors. The sensor is capable of detecting the variation on the current produced when toxic compounds (e.g., formaldehyde) are present in the medium. Stein et al. (2011) modified a bioelectrochemical model combined with enzyme inhibition kinetics, which describes the polarization curve of an MFC-based biosensor, to describe four types of toxicity. They discovered that the overpotential has to be controlled in order to get a stable and sensitive biosensor. Based on the knowledge gained, Stein et al. (2012) experimentally investigated the sensitivity of a MFC-based biosensor for nickel. They found that the effect of four types of ion exchange membranes (cation exchange, anion exchange, monovalent cation exchange and bipolar membranes) on the sensitivity was not significant. The sensor had higher response at high overpotentials, even nickel concentration is low.

3.3 Sediment MFC as power sources or bioremediation

A potential application of MFC technology is in remote marine or freshwater environments where electricity can be harvested from organic-rich aquatic sediments (He et al., 2007; Paper V; Paper VI). The above process is always carried out in a so-called sediment MFC system, as shown in Figure 3. The benefits of sediment MFC go beyond energy generation. Sediment MFC can

enhance the oxidation of organic matters at the anode, thus bringing about the removal of excessive or unwanted pollutants from sediments. Furthermore, sediment MFC could also be applied to accelerate cycle of some metals such as manganese (De Schamphelaire et al., 2008a; Rhoads et al., 2005).

3.3.1 Development

The first sediment MFC was demonstrated by Reimers et al. (2001). By embedding pairs of platinum mesh or graphite fiber-based electrode in marine sediment, the other in seawater, they have observed 10 mW/m^2 of power density from marine sediment-seawater interfaces. One year later, Bond (2002) report a specific enrichment of microorganisms of the family *Geobacteraceae* on energy-harvesting anodes, and found that these microorganisms can oxidize organic compounds to conserve energy and support their growth by using electrode as the sole electron acceptor. Their finding not only offers an approach for harvesting energy from organic-rich sediments, but also provides a new bioremediation technology for organic pollutants removal from subsurface environments.

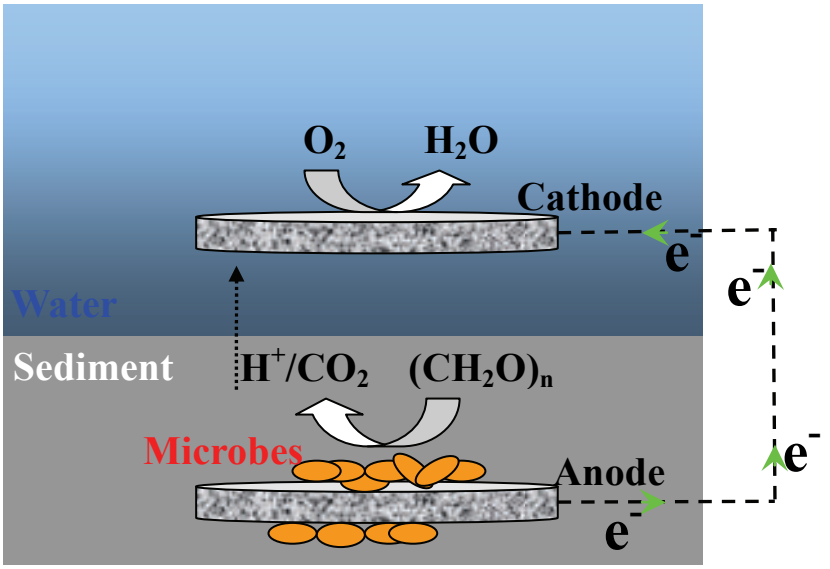

Figure 3. Schematic diagram of typical sediment MFC

3.3.2 System improvements from anode side

Though sediment MFC shows promising perspective in energy production and bioremediation, the power generations from this system are still low, which is around 10-20 mW/m^2 at its earliest development (Reimers et al., 2001; Tender et al., 2002). Efforts are being made to improve the power generation and voltage

output. It was realized that the power generation significantly depends on the level of organic matter in sediment, oxygen availability in the overlying water, electrode design and spacing, external resistance, conductivity and temperature of water (Hong et al., 2009; Reimers et al., 2001; Ryckelynck et al., 2005; Tender et al., 2002; Paper V). Lowy et al. (2006) observed that power densities from sediment with organic contents of 4-6% can be increased up to 2.5 times by modifying the anode with different metals or known mediators. For example, 98 mW/m^2 was produced by bonding AQDS (9,-10-anthraquinone-2,6-disulfonic acid) on the anode, and 105 mW/m^2 was produced by using ceramic-graphite composite anodes containing Mn^{2+} and Ni^{2+}. However, power generations in above study are not sustained and decreased rapidly over time due to the depletion of metals or mediators. To increase power generation from the sediments with low organic content, biodegradable particulate substrates including Chitin 20, Chitin 80 and cellulose power were added to the anode compartment and a maximum power density of 84 mW/m^2 was produced (Rezaei et al., 2007). However, this approach also has its own drawbacks such as the limited half-life and poor mass-transfer in the sediment matrix (Reimers et al., 2001).

3.3.3 System improvements from cathode side

Efforts have also been made to increase the cathode design and oxygen availability. He et al. (2007) developed a sediment MFC with a rotating cathode to increase the oxygen availability to the cathode, and they observed a higher power production of 49 mW/m^2 compared to 29 mW/m^2 in a nonrotating cathode system. Sustained and high voltage outputs were obtained by using brush cathodes containing graphite carbon fibers instead of spinal coated stainless steel wool cathodes because a 50% reduction of internal resistance (Hasvold et al., 1997). Biofilm colonized biocathode was also employed to catalyze the cathodic oxygen reduction and accordingly increase the power generation from sediment MFCs (Chen et al., 2012b; De Schamphelaire et al., 2010).

3.3.4 Power sources

With the improvement of power generation, sediment MFCs are considered to be an alternative renewable power source to conventional batteries for remote devices used in freshwater and marine environments (e.g., benthic sensors, telemetry systems) (Dewan et al., 2010; Donovan et al., 2008; Paper V). Using sediment MFC as viable power supply was firstly demonstrate by Tender et al.

(2008). They successfully powered a meteorological buoy with a sediment-MFC constructed in a marine environment. To power high-power devices, the electricity produced by a sediment MFC could be firstly stored in a capacitor, and then be further transferred to the powering devices. Donovan et al. (2008) developed a sediment MFC, producing continuous power between 1 and 4 mW in the winter and summer, respectively, to power a wireless sensor requiring 11 mW. In order to power the wireless sensor, they first stored the electric energy in a capacitor and using it intermittently. When the capacitor potential reached 320 mV the wireless sensor was powered. Instead of external resistor, Dewan et al. (2010) evaluated the performance of sediment MFC using an energy-storing device such as a capacitor, and their system was further used to power wireless sensors monitoring the environment. Shantaram et al. (2005) previously used an MFC with a microbial cathode and a sacrificial anode to power a wireless temperature sensor. Their configuration increased the cell potential above 1.2 V to power a sensor with appropriate off-the-shelf electronics. However, use of sacrificial anodes limits the lifespan of their system. In order to improve the power generation of sediment MFC as a power sources, Zhang et al. (2011) arranged cathodes in two different way, floating cathode and bottom cathode. Higher power was produced from the system with a floating cathode, which resulted in a shorted charging time when an ultracapacitor is connected to the circuit. A power management system was developed to boost the voltage (0.5 V) to 3.3 V for powering a wireless temperature sensor.

3.3.5 Bioremediation

Besides power sources, sediment MFCs have also been employed as a bioremediation technology (Hong et al., 2010; Morris and Jin, 2012; Paper V). It has been found that the sediment organic matter (SOM) was microbially oxidized under anaerobic conditions with an electrode serving as a terminal electron acceptor. Compared with original sediment, SOM around the electrodes became more humified, aromatic, and polydispersed, and had a higher average molecular weight (Hong et al., 2010). These findings may present a potential for the energy-efficient remediation. Sediment MFC has been employed to remove various organic pollutants from subsurface environments, such as phenanthrene and pyrene in freshwater sediment, uranium-contaminated aquifers, aromatic hydrocarbon-contaminated sediments and 1,2-dichloroethane (Huang et al., 2011; Morris and Jin, 2012; Pham et al., 2009; Williams et al., 2010a; Williams et al., 2010c; Yan et al., 2010; Yuan et al., 2010)

3.4 H_2 production: microbial electrolysis cell (MEC)

3.4.1 Definition and working principle

High-efficiency biological hydrogen production can be realized by a technology termed MEC (Cheng and Logan, 2007b; Liu et al., 2005c; Logan et al., 2008; Paper II). The MEC is developed on the base of MFC and they share many common characters such as similar reactor design and anode reaction (Logan et al., 2008; Logan et al., 2006). An MEC can be easily switched from an MFC by excluding oxidants (e.g., O_2) from the cathode and providing a certain amount of electricity. A typical two-chamber MEC is shown in Figure 4. MEC has similar anodic oxidization catalyzed by microorganisms as MFC. The main differences are that MEC requires electricity supply and end product is hydrogen instead of electricity. Moreover, MEC cathode is in anaerobic condition while MFC cathode requires aeration. In the anode of MEC, bacteria oxidize organic matters and release protons, electrons and carbon dioxide; the protons transfer across the membrane and reach the cathode of MEC. If there is no external voltage supply, hydrogen production is not spontaneous. However, if an external potential is applied in this situation between the anode and cathode, hydrogen gas is produced at the cathode by reduction of protons (Cheng and Logan, 2007b). Theoretically, an applied voltage of 0.14 V is required for hydrogen production in the condition that acetate is used as substrate. But usually a voltage higher than 0.2 V should be applied due to overpotentials at the electrodes (Rozendal et al., 2006b).

3.4.2 Advantages

MECs have two main advantages over other biological hydrogen production processes. First, various organic matters can be used as substrate, including cellulose, glucose, glycerol, acetic acid, sewage sludge and varied wastewaters (Cheng and Logan, 2007a; Liu et al., 2005c; Logan et al., 2008). Second, non-fermentable substrates (e.g., acetate, butyrate), which are byproducts of dark fermentation due to thermodynamical limitations, can be completely oxidized, resulting in high H_2 yields (Liu et al., 2005c; Logan et al., 2008). MEC can theoretically produce 12 mol H_2/mol glucose compared to the maximum 4 mol H_2/mol glucose produced in conventional fermentation process. Unlike typical water electrolysis (>2.1 V), MEC technology requires relatively low energy input (0.2-0.8 V) which is could be another advantage.

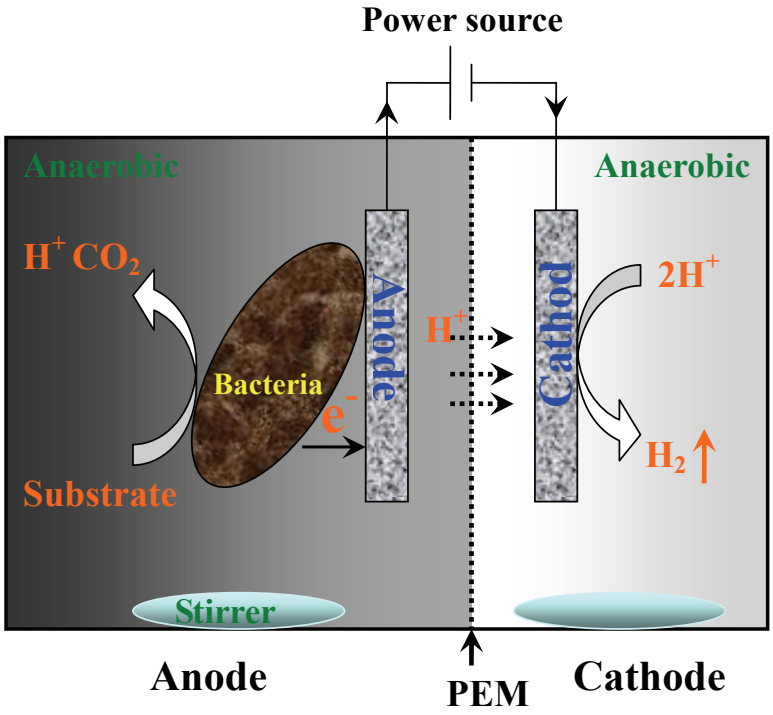

Figure 4. Schematic diagram of typical two-chambered MEC

3.4.3 Research focuses

MEC technology is still in its infancy and has not been removed from bench scale operation. Since MEC is developed on the base of MFC and shares most of the common limitations with MFCs, thus the advances in MFCs could also be applied to MEC. Similar as MFCs, efforts are being made to improve the system performance, construction and operation cost. The research interests are mainly lie on potential substrates, new reactor design, electrode material and modification, separator materials, methanogens inhibition and integration with other technologies.

Substrate type and concentration are important factors affecting the performance of MFC, which are also applied to MECs. Nearly all substrates used in MFC could also be used in MECs. Until now, several organic compounds from pure chemicals such as acetate, butyrate, propionate, glucose, lactic acid, valeric acid, glycerol, cellulose to even complex waste streams such as protein, domestic wastewater, industrial wastewater, winery wastewater and waste activated sludge have been tested as substrate of MECs (Cheng and Logan, 2007b; Cusick et al., 2010; Liu et al., 2005c; Lu et al., 2012a; Lu et al., 2012b; Lu et al., 2010; Pant et al., 2012). In a certain range, relatively higher substrate concentration results in higher hydrogen production (Paper II).

Reactor design is one of the most important affecting factors. Generally, most of the MFC reactor designs can be modified for electricity-assisted biohydrogen production. Several different architectures have been tested. The earliest systems are two-chambered system such as H-type reactors, which were designed only for "proof of concept" and thus the performance was not optimized (Liu et al., 2005c). There are some two-chambered systems used in fed-batch experiments, such as two cube-type MECs (512 and 42 mL, respectively) where anode and cathode are separated by a membrane (Cheng and Logan, 2007b; Ditzig et al., 2007). There are also some two-chambered systems used in continuous flow tests, such as disc-shaped two-chamber MEC, disk-shaped membrane electrode assembly MEC with gas diffusion electrode, and rectangular-shaped MEC with serpentine-shaped flow channels through the reactor that allow the gas to be released at the top of each flow path (Rozendal et al., 2006b; Rozendal et al., 2007; Rozendal et al., 2008b; Rozendal et al., 2008c). Compared with two-chambered system, single-chamber MECs seems more cost-effective. The typical single chamber MEC is modified from the single-chamber air cathode MFC (Call and Logan, 2008; Hu et al., 2008). Hu et al. (2009) developed a single-chamber tubular MEC using non-precious-metal catalysts. In Paper II, an innovative reactor design termed submersible self-powered MEC was developed, which offers insight into cost-effective reactor design of MEC system.

Separator and its impact is well studied in MFCs (Liu and Logan, 2004; Logan et al., 2006), while it is also important to the performance of MECs. The separators used in MECs are generally CEMs (Rozendal et al., 2007). The appearance of membrane is essential for the purity of the hydrogen, but it may increase internal resistance. Removing the membrane from MEC may reduce the internal resistance and construction cost but may lower the Coulombic efficiency and the produced hydrogen could be polluted by the off-gases from anode or even consumed by methanogens (Call and Logan, 2008; Rozendal et al., 2007). To reduce the internal resistance caused by large electrode spacing, membrane cathode assembly has also been used in MECs (Jia et al., 2012). In Paper II, a sandwich type membrane electrode assembly was used to enhance the hydrogen production in a so called self-powered submersible MEC.

Electrodes materials used in MFCs could principally be used in MECs. The hydrogen evolution rate (HER) is strongly dependent on the cathode electrode materials. Catalyst such as platinum has often been used to decrease the overpotential and increase the kinetics of HER (Logan et al., 2008). However,

platinum is expensive metal and not suitable for practical application. Some cost-effective and efficient electrodes and catalysts that have been tested in MFCs have also been applied in MECs, such as tungsten carbide, various stainless steel, nickel alloys (Call et al., 2009; Harnisch et al., 2009; Logan, 2010; Selembo et al., 2009). Beside metal catalyst, biocathode that catalyzed biohydrogen production has been developed and showed good performance (Rozendal et al., 2008b).

Methanogenesis is commonly observed in the anode of MECs due to the growth of methanogens. The amount of methane produced from MEC reactors are varied with inoculum, substrate and reactor configuration (Chae et al., 2010). The appearance of methanogens is not expected in MECs, as it will reduce the hydrogen production and purity, and thus result in energy losses. This suggests that the specific method for suppressing methanogens is critical for improving the overall hydrogen recovery in MECs. Several strategies for inhibition of methanogenesis in MECs have been suggested, such as periodical exposure to air, operation under low pH, washout of methanogens by a short hydraulic retention time and low substrate concentrations (Call and Logan, 2008; Clauwaert and Verstraete, 2009; Rozendal et al., 2008b). However, most of methods are not efficient or expensive for practical operation. In Paper II, alternate changing the hydrogen-producing cell to electricity-assisting cell was proposed to inhibit methane production. Compared with typical air exposure method, this approach has no negative impact on the system.

3.5 Desalination: microbial desalination cell (MDC)

3.5.1 Definition and working principle

A new technology for seawater desalination was recently developed on the base of MFC, called MDC. The working principle of MDC is on the base of a phenomenon that ion transport is needed to maintain charge balance in the anode and cathode of MFCs (Rozendal et al., 2006a). The main difference of MDC compared with MFC is that MDC contains an additional desalination chamber between anode and cathode chambers separated respectively by AEM and CEM (Cao et al., 2009). In a typical three-chambered MDC system (Figure 5), when electrochemically active bacteria oxidize organic matters and transfer electrons to the external circuit, protons are released into solution and accumulated in the anode, the AEM deposited in the anode side prevents positively charged species from leaving the anode and thus negatively charged species move from the

middle chamber to the anode to maintain charge balance. In the cathode chamber, protons are consumed to form water with electrons, resulting in the movement of positively charged species from the middle chamber to the cathode chamber (Cao et al., 2009). In such way, the salt ionic species are removed from the middle chamber results in water desalination.

3.5.2 The main advantages

Compared with conventional desalination technologies, in MDC, salt was removed without any water pressurization or use of draw solutions, and no electrical energy is required. Instead, electricity production is accomplished with water desalination (Cao et al., 2009; Jacobson et al., 2011b).

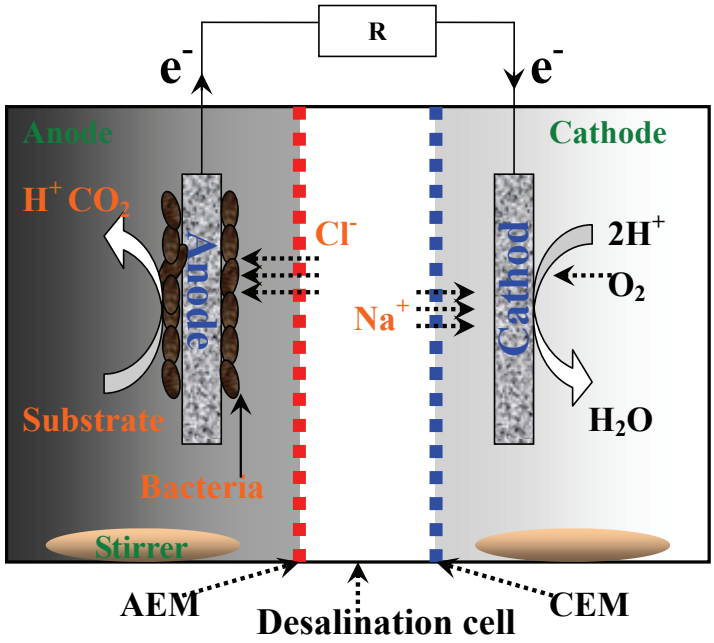

Figure 5. Schematic diagram of typical MDC

3.5.3 Research focuses

Three-chamber system-The MDC concept was successfully demonstrated by Cao et al.(2009) using a ferricyanide catholyte in a three chamber system. To make this system more efficient and to replace the toxicity ferricyanide with oxygen at cathode, Mahanna et al. (2010b) developed a new type of air-cathode MDC containing three equally sized chambers. They found that it is possible to remove up to 60% of the salt using equal volumes of water in the anode and desalination chamber. In this system, up to 480 mW/m^2 of power density was produced and no external power source was needed. It has been found that the accumulation of proton in three-chamber MDC can lead to pH variation and

affect the system performance. Qu et al. (2012) developed a new type of modified MDC, called a recirculation MDC (rMDC) to reduce pH variations in the electrode chambers and increase power densities.

Two-chamber system-To further reduce the construction cost, Jacobson et al. (2011a) eliminated the cathode chamber and developed a continuously operated upflow MDC (UMDC) for the purpose of salt removal. Later, the same researchers developed their UMDC to the liter-scale (total volume of 2.75 L) and used this L-scale UMDC to study desalination with either salt solution (NaCl) or artificial seawater (Jacobson et al., 2011b). Such reactor design indeed offers insight into reduction of the construction cost and reactor scale-up, but using acidified water as catholyte to rinse the cathode may add the operation cost. In Paper VII, an innovative two-chamber MDC was developed.

Stacked MDCs were developed in order to promote the desalination rate. Chen et al. (2011) developed a stacked MDC by using several pairs of AEM and CEM to separate the anode and cathode, resulting in several desalination chambers. The total desalination rate was indeed increased in such system, but increase of the number of desalination chamber also raised the internal resistant and resulted in low salt removal efficiency. To solve above problems and improve the system performance from a stacked MDC, Kim and Logan (2011) developed a stacked MDC using thin stacks, air cathode and novel water flow scheme. The performance of this system was greatly improved compared to previous MDC with one desalination chamber or two cell-paired MDC stack.

Integration with other processes is another research focus of MDC. One main limitation of the MDC reported so far is that the voltage is not constant over the desalination. An alternative approach is to boost the voltage produced by the bacteria using a power source and result in hydrogen production at the cathode (Luo et al., 2011; Mehanna et al., 2010a). Such process is kind of integration of MDC and MEC technologies, which can concurrently desalinate salt water, produce hydrogen, and potentially treat wastewater. Chen et al. (2012a) recently developed a microbial electrolysis desalination and chemical production cell with four chambers using a bipolar membrane. To supply a certain amount of voltage to this system, the production of acid (HCl) and alkali (NaOH) was accomplished with seawater desalination.

3.6 Other possibilities

MFC-based technology, such as microbial electrosynthesis, can be applied for production of fuels and chemicals. The key examples are production of ethanol, methane, H_2O_2, caustic soda (Rabaey et al., 2010; Rabaey and Rozendal, 2010; Rozendal et al., 2009; Steinbusch et al., 2010; Wagner et al., 2009).

4 Improvement of MFC-based technologies

According to above sections, to improve the performance and reduce the construction cost for different MFC-based technologies, main research interests are lay on reactor design, expanding the scope of application, integration with existing technologies, which are also the focuses of this study.

4.1 Integration with other technologies

MFC and its based technologies have been proposed as independent systems for harvesting energy from waste. However, MFC-based technologies are still in its "infancy", in some case, they can not compete with conventional technologies in energy production. Thus, MFC-based technologies can be a pretreatment or down-stream technology for some conventional technologies. For example, MFCs has been proposed as supplement of anaerobic digestion (Pham et al., 2006). It has also been proposed that MDC technology could be as partial process of reversal osmosis (Mehanna et al., 2010b). Integration MFC-based technologies with conventional technologies may bring the benefits from each technology together, and thus improve treatment efficiency and save energy cost.

In paper I, a new concept based on the integration of MFC and microalgae technology was proposed to reduce the cost and expand the application of MFCs in wastewater treatment. Through this synergistic cooperation, more than 99.6% of C, 87.6% of N and 69.8% of P was removed simultaneously from wastewater without extra cost (e.g. mechanical aeration), while electricity and valuable algae biomass were produced simultaneously.

In paper VII, a newly designed MDC system was integrated with electrochemical denitrification for nitrate removal from groundwater. Through this integration, nitrate can be *in situ* removed from groundwater without energy input and the risk of bacteria discharge.

4.2 Cost-effective and *in situ* applicable reactor design

It has been recognized that improvement of the reactor design could significantly contribute to overcome some of the present limitations. To improve the system performance and reduce the construction, operation and maintenance cost in different applications, different reactor designs may be required (Clauwaert et al., 2008; Logan, 2010; Logan et al., 2006). For example, most of the existing MFC

sensors are surface applicable systems (Di Lorenzo et al., 2009a; Kim et al., 2003; Kumlanghan et al., 2007; Liu and Mattiasson, 2002), no information about their application in subsurface environments (e.g., groundwater) is available. Thus *in situ* applicable configuration is important for biosensors especial the one to be applied to existing anaerobic reactors or subsurface environments (e.g., groundwater, sediment).

In Paper III, a biosensor (SUMFC), based on a submersible microbial fuel cell, was developed for *in situ* monitoring of microbial activity and BOD in groundwater. With this design, specially designed anode chamber was not needed, which makes it easy to apply in groundwater and other natural anaerobic environments, thereby avoiding the cost for extensive sampling, pumping and distribution.

In Paper IV, a submersible microbial fuel cell sensor (SBMFC) was developed for *in situ* and real time monitoring of DO in environmental waters. The SBMFC has only anode chamber, which makes it applicable for *in situ* monitoring of various environmental waters.

In Paper II, a self-powered submersible microbial electrolysis cell (SMEC), in which a specially designed anode chamber and external electricity supply were not needed, was developed for in situ biohydrogen production from anaerobic reactors. The simple, compact and in situ applicable SMEC offers new opportunities for reactor design for a microbial electricity assisted biohydrogen production system.

In Paper V, an innovative self-stacked submersible MFC (SSMFC) was developed to improve the voltage generation from lake sediments. The SSMFC, composed mainly of one chamber and two pieces of sandwich-type electrode, could function as a two-cell stacked MFC with reduced electrode spacing, thereby improving voltage output. Moreover, a dedicated anode chamber was not required in such SSMFC design, which makes it *in situ* applicable to natural anaerobic environments, such as sediments.

In Paper VII, a submersible MDC, composed of only two chambers, was developed for nitrate removal from groundwater. In such design, a desalination chamber which is indispensable part of traditional MDC is not required. The reactor can be submersed in groundwater, seawater or brackish reservoir.

4.3 Expanding the scope of application

As stated in section 3, the performance of most MFC-based technologies are relatively low compared to conversion technologies, thus the application field of MFCs should have to be shifted from electricity production toward to specialized value-added applications.

In Paper I, the integration of MFC and microalgae cultivation has expanded the application of MFC from organic matters and nitrogen species removal to simultaneous C, N and P removal.

In Paper IV, using of submersible MFC as a DO sensor has expanded the application of MFC sensor from BOD, microbial activity and toxicity detection to DO monitoring.

In paper VII, the application of a novel MDC system was expanded from water desalination to nitrate removal from groundwater.

5 Conclusions

This thesis has mainly focused on improvement of the performance, reduction of the construction, maintenance and operation costs, and expanding application field for different MFC-based systems and applications. The major contributions of this thesis work are summarized as follows:

- ★ A novel wastewater treatment process, based on the integration of MFC and microalgae cultivation technologies, was developed to produce electric energy and valuable biomass, and remove carbon, nitrogen and phosphorus from wastewater simultaneously. Under illumination, stable power density of 68±5 mW/m^2 and biomass of 0.56±0.02 g/L were generated at initial algae concentration of 3.5 g/L. Accordingly, the removal efficiency of organic carbon, nitrogen and phosphorus was 99.6%, 87.6% and 69.8%, respectively. The main removal mechanism of nitrogen and phosphorus was algae biomass uptake. The system performance was affected by illumination period, algae concentration and the ratio of carbon and nitrogen.

- ★ An *in situ* applicable SUMFC biosensor was developed for monitoring of microbial activity and BOD in groundwater. The sensor using fresh anode was excellent for monitoring microbial activity, whereas, the sensor using biofilm-colonized anode was appropriate for BOD measurement. BOD of up to 250 mg/L and the concentration of active microorganisms up to 6.52 nmol-ATP/L can be measured based on a linear relation with current density, respectively. The sensitivity of the sensor was affected by operation conditions such as temperature, initial pH, and conductivity. The sensor showed good performance with real groundwater samples and a deviation less than 22% was observed.

- ★ The application of MFC-based sensor was expanded for DO monitoring in environmental waters. SBMFC was developed as *in situ* and real time biosensor. DO levels up to 8.8±0.3 mg/L can be detected with a maximum response time of 4 min. The external resistance and anodic substrate concentration affected the current density. The sensor performance was also influenced by environmental conditions of the monitored waters, including temperature, pH, conductivity and alternative electron acceptors.

★ A SMEC, in which a specially designed anode chamber and external electricity supply were not required, was developed for in situ biohydrogen production from anaerobic reactors. The hydrogen production increased with acetate and buffer concentration. The highest hydrogen production rate of 32.2 mL/L/d and hydrogen yield of 1.43 mol-H_2/mol-acetate were achieved with20 mM acetate and 100 mM phosphate buffer. Alternate exchanging the function between the cells in the SMEC was found to be an effective approach for inhibiting methanogens. The Cell 1 and Cell 2 in the SMEC had the same anodic microbial community composition. The SMEC with simple and compact configuration showed promising perspective for *in situ* hydrogen production from various anaerobic environments by directly utilizing the electricity produced from waste.

★ A SSMFC was developed to improve the voltage output from lake sediments. This is the first attempt to improve the electricity generation from sediment by employing stack operation. The SSMFC successfully produced a maximum power density of 294 mW/m^2 and had an OCV of 1.12 V. The voltage reversal, which is a major limitation in MFCs during stack operation, was studied detail in terms of cause, affecting factors and elimination methods.

★ A sediment-type MFC based on two pieces of bioelectrodes was employed as a novel *in situ* applicable approach for nitrogen removal, as well as electricity production from eutrophic lakes. A laboratory system was employed to prove the concept. Power density of 42 and 36 mW/m^2 were produced respectively from nitrate and nitrite synthesized lake waters, while 62% and 77% of total nitrogen removal were accomplished. The nitrogen removal in the system was almost 4 times higher compared to the open-circuit operation. The results provide an insight into the bioremediation of nitrogen polluted surface waters in a cost-effective and sustainable way.

★ An innovative submersible microbial electrodialysis-denitrification cell (SMDDC) was development as an *in situ* applicable technology for removing nitrate from groundwater. The system performance was mainly determined by the nitrate concentration and ionic strength of groundwater. Meanwhile, improved system performance and minimized

electron losses were achieved with well controlled anodic HRT and low external resistance. The SMDC showed high nitrate removal and current generation with high ionic strength of groundwater and was able to reduce the salinity of groundwater as well. External nitrification of the anodic effluent was beneficial to the current generation and nitrate removal rate, but was not helpful for total nitrogen removal. This technology offers a new avenue to remove nitrate from subsurface environments such as groundwater.

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7 Papers

I. Zhang Y., Noori J.S., Angelidaki I. 2011. Simultaneous organic carbon, nutrients removal and energy production in a photomicrobial fuel cell (PFC). *Energy and Environmental Science* 4(10): 4340-4346.

II. Zhang Y., Angelidaki I. 2012. Innovative self-powered submersible microbial electrolysis cell (SMEC) for biohydrogen production from anaerobic reactors. *Water Research* 46(8): 2727-2736.

III. Zhang Y., Angelidaki I. 2011. Submersible microbial fuel cell sensor for monitoring microbial activity and BOD in groundwater: Focusing on impact of anodic biofilm on sensor applicability. *Biotechnology and Bioengineering* 108(10):2339-2347.

IV. Zhang Y., Angelidaki I. 2012. A simple and rapid method for monitoring dissolved oxygen in water with a submersible microbial fuel cell (SBMFC). *Biosensors and Bioelectronics* 38(1):189-194.

V. Zhang Y., Angelidaki I. 2012. Self-stacked submersible microbial fuel cell (SSMFC) for improved remote power generation from lake sediments. *Biosensors and Bioelectronics* 35(1):265-270.

VI. Zhang Y., Angelidaki I. 2012. Bioelectrode-based approach for enhancing nitrate and nitrite removal and electricity generation from eutrophic lakes. Moderate revision in *Water Research.*

VII. Zhang Y., Angelidaki I. 2012. A new method for *in situ* nitrate removal from groundwater using submersible microbial desalination-denitrification cell (SMDDC). *Submitted manuscript.*

The papers **I-VII** are included in the printed version of the thesis but not in the www-version.
Copies of the papers can be obtained from the
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Department of Environmental Engineering
Technical University of Denmark
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(library@env.dtu.dk)

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Water Resource Engineering, Urban Water Engineering,
Residual Resource Engineering and Environmental Chemistry & Microbiology.

The department dates back to 1865, when Ludvig August Colding, the founder of the department, gave the first lecture on sanitary engineering as response to the cholera epidemics in Copenhagen in the late 1800s.

DTU Environment
Department of Environmental Engineering
Technical University of Denmark

Miljoevej, building 113
DK-2800 Kgs. Lyngby
Denmark

Phone: +45 4525 1600
Fax: +45 4593 2850
e-mail: reception@env.dtu.dk
www.env.dtu.dk

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